- Abstract -

HANFORD RPP-WTP HIGH-LEVEL WASTE VITRIFICATION OFFGAS SYSTEM

James K. Rouse, P.E.

The United States Department of Energy (DOE) and CH2M Hill Hanford Group are currently designing a facility for the pretreatment and vitrification of radioactive waste stored in underground storage tanks at the Hanford Site in Washington State. This project will soon enter the detailed design phase. The waste retrieved from these tanks will be conditioned in a pretreatment facility. The high-level waste fraction will be sent to a vitrification facility for immobilization in glass by a Joule heated ceramic melter. The vitrification process offgas will be treated to assure compliance with regulations prior to release.

The offgas from the high-level waste (HLW) vitrification melter is generated by and composed of the following:

- Air from melter bubbler operation and inleakage into the melter,
- Water vapor evaporated from the melter feed,
- Acid gases generated from feed decomposition (i.e., CO_2, NO_x, SO_x, etc.), and
- Aerosols from dried melter feed and melter cold-cap reaction solids.
- Air is also added to the melter offgas from the follow:
  - Air Inleakage into the Melter
  - Melter Bubblers
  - Film Cooler Air Inbleed
  - Control Air Added to Regulate Melter Plenum Vacuum

In addition, the HLW melter also generates small quantities of other volatile compounds that may require attention during treatment. These compounds include iodine-129, nitrous oxide compounds, and/or volatile organics. The vessel ventilation offgas is added to the melter offgas during this treatment and discharged together.

Offgas treatment systems are provided to remove aerosols, acid gases, and radionuclide contaminates. The system components include:

- Primary HLW Melter Offgas Treatment System
  - Film Cooler
  - Submerged Bed Scrubber (SBS)
  - Wet Electrostatic Precipitator (WESP)
  - High Efficiency Mist Eliminator (HEME)
  - High Efficiency Particulate Air (HEPA) Filter
- Secondary Offgas Treatment System
  - Packed Bed Caustic Scrubber
  - Volatile Organic Compounds Thermal Catalytic Oxidizer

The vessel ventilation air is combined with the WESP offgas discharge. The combined offgas is further treatment by the primary HEMEs, HEPAs, and the secondary offgas system. After treatment by the secondary offgas treatment system, the combined HLW offgas is discharged to the facility main stack.

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Introduction

The United States Department of Energy (DOE) and its contractors manage 177 underground radioactive waste storage tanks at the Hanford Site in Washington State. These 177 tanks currently contain approximately 54 million gallons of radioactive waste. This waste is comprised of 14 million gallons of sludge (metal precipitates), 24 million gallons of "saltcake" (e.g., primarily dried sodium nitrate and nitrite), and 16 million gallons of radioactive liquid waste or "supernatant."

It is the intention of DOE to remove this radioactive waste from the underground tanks and provide it as feedstock to the River Protection Project – Waste Treatment Plant (RPP-WTP). This waste will be separated into Low-Active Waste (LAW) and High-Level Waste (HLW) fractions. The HLW fraction will consist of the insoluble tank sludge as well as the technetium, transuranics, and cesium removed from the LAW fraction in the Pretreatment facility. Some supernatant containing soluble species will also be present.

Two vitrification systems are being designed to immobilize the radioactive waste: one for LAW feed and the second for HLW feed. Waste will be immobilized in the form of glass monoliths in metal canisters. Canisters containing immobilized HLW and LAW will be temporarily stored at the RPP-WTP and then transferred to permitted storage or disposal facilities.

HLW Vitrification Offgas Treatment System Description

Vitrification Process

The purpose of the HLW vitrification off-gas treatment system is to remove aerosols and noxious gases from the HLW melter off gas and the vessel ventilation system. Melter off gas is generated from the vitrification of HLW slurry, which is feed to a joule-heated ceramic melter. A single HLW melter will initially be operated at approximately 1.5 metric tons per day. Early pilot plant tests show that this production rate could be ramped up to 3.0 Mt/day. Space for a second future HLW melter will be available within the HLW facility. A separate off gas treatment system will be installed for each melter. After treatment, this off gas will be discharged through the HLW vitrification facility main stack. Figure 1 provides a simplified representation of a vitrification melter in operation.

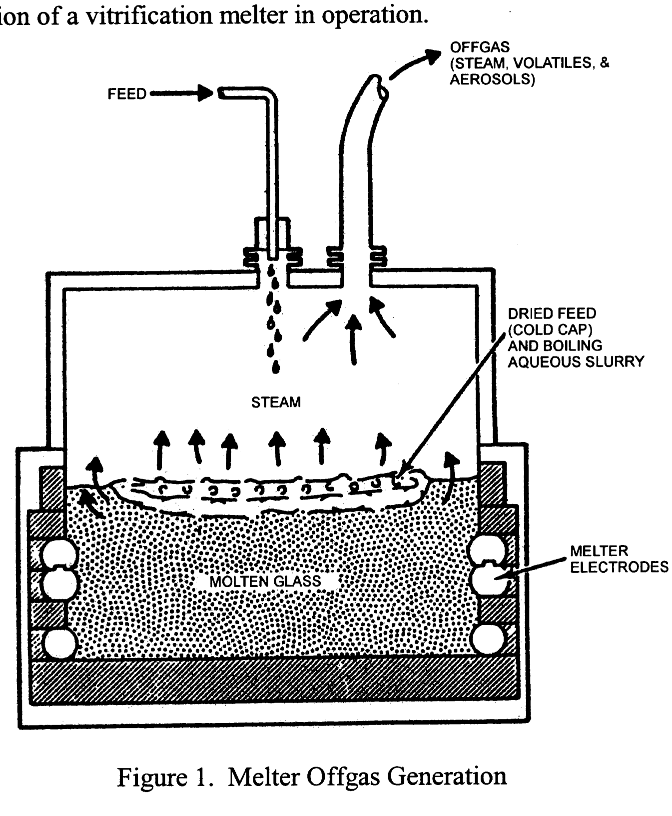

Figure 1. Melter Offgas Generation

System Challenges/Melter Off Gas Description

The principal components of the HLW melter off gas stream will be air and steam. The offgas air component will be derived from four major sources as follows:

- Melter molten glass air sparging and instrumentation
- Air inleakage into the HLW melter
- Air injection into the melter film cooler
- Melter pressure control air inbleed
- Steam from water vapor evaporated from the melter feed
- Vessel and Pulse ventilation off gas

Melter air sparging, instrumentation air, and air injection into the film cooler are assumed to be constant during melter operation. Air inleakage into the HLW melter will vary based on the changing pressure depression between the melter plenum and the melter cell. Pressure control air will be added to the off gas stream after the film cooler. It will vary accordingly to maintain a constant vacuum in the melter plenum. Water present in the melter feed will vaporize to steam.

Secondary components of the HLW off gas stream are as follows:

- Acid gases generated from anion decomposition (i.e., CO_2, NO_x, SO_x, etc.),
- Aerosols from dried melter feed and melter cold-cap reaction solids

Acid gases are generated from the decomposition of salts and organic compounds present in the feed. Aerosols are present due to cold cap solids being entrained in the off gas stream. The aerosols will be bimodal in size distribution, having a significant fraction larger then 16 microns and another significant fraction less then 1.6 microns. The large particulates are from cold cap solids entrainment and the smaller aerosols from the condensation of semi-volatile off gas components.

Treatment Equipment Selection

The current HLW off gas treatment flowsheet was selected through an evaluation of melter off gas systems in operation around the world. This included the melter off gas systems at the following locations:

- West Valley Nuclear Services, New York
- Defense Waste Processing Facility, Savannah River Site, South Carolina
- Vitrification Facility, Sellafield, England
- Tokai-Mura, Japan
- Pamela, Belgian

Six different off gas treatment flowsheets were prepared that were roughly based on these existing systems, but also taking into account the unique needs of the RPP-WTP HLW melter. Individual off gas treatment technologies that were evaluated included the following:

- Submerged Bed Scrubbers
- Ejector-Venturi Scrubbers
- Hydrosonic Scrubber
- Wet Electrostatic Precipitators
- High Efficiency Mist Eliminators
- High Efficiency Particulate Air Filters
- High Efficiency Metal Filters
- Pack Bed Caustic Scrubbers
- Silver Mordenite Absorption Columns

Each flowsheets was evaluated based on overall capital and operational cost, overall collection efficiency for particulates and gases, perceived operability and ease of maintenance, and a hazard analysis performed over all six systems. The current system is a product of this evaluation.

Treatment Equipment Description

The HLW vitrification off gas treatment system consists of two subsystems, primary and secondary. The primary offgas system is designed to quench the melter off gas and to remove entrained aerosols. Essentially all radionuclides are removed, with the exception of iodine-129. The secondary off gas system is designed to remove specific compounds, specifically volatile organic compounds, NO_x species, and iodine-129. Overviews of these two subsystems are illustrated in Figures 2 and 3, respectively. Treatment equipment within these subsystems are defined as follows:

- Primary HLW Melter Off Gas Treatment System
  - Film Cooler
  - Submerged Bed Scrubber (SBS)
  - Wet Electrostatic Precipitator (WESP)
  - High Efficiency Mist Eliminator (HEME)
  - High Efficiency Particulate Air (HEPA) Filter
- Secondary Off Gas Treatment System
  - Packed Bed Caustic Scrubber
  - Volatile Organic Compound Thermal Catalytic Oxidizer

The HLW melter off gas is initially cooled and accelerated through the melter film cooler. The film cooler is essentially a double-walled pipe designed to mix process air with the melter off gas. Compressed air is introduced axially through a series of slots or holes in the inner wall of the film cooler. It then mixes with the melter off gas, allowing it to cool from its melter plenum temperature of ~400°C to a discharge temperature between 250 and 350 °C. This facilitates the cooling of the off gas below the glass "sticking" temperature to minimize solids deposition on off gas piping walls. The injection of compressed air into the melter off gas also helps maintain an off gas flow velocity of about 60 ft/s. This gas velocity has also been found to minimize solids build-up within the film cooler.

The off gas from the HLW melter film cooler is further cooled and treated by a submerged bed scrubber (SBS). The SBS is a passive device designed for the aqueous scrubbing of entrained radioactive aerosols from the melter off gas. It will also cool the off gas to a desired discharge temperature through the use of coiling coils. The off gas leaves the SBS in thermal equilibrium with the scrubbing solution. As shown in Figure 4, the off gas enters the SBS through an inlet pipe that runs down through the center of the bed to the packing support plate.

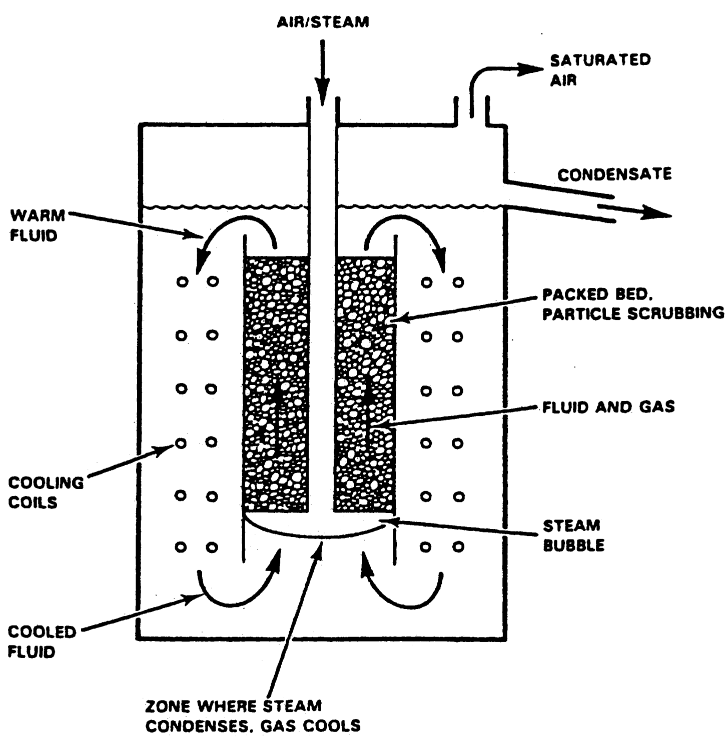

Figure 4. Submerged Bed Scrubber (Schematic)

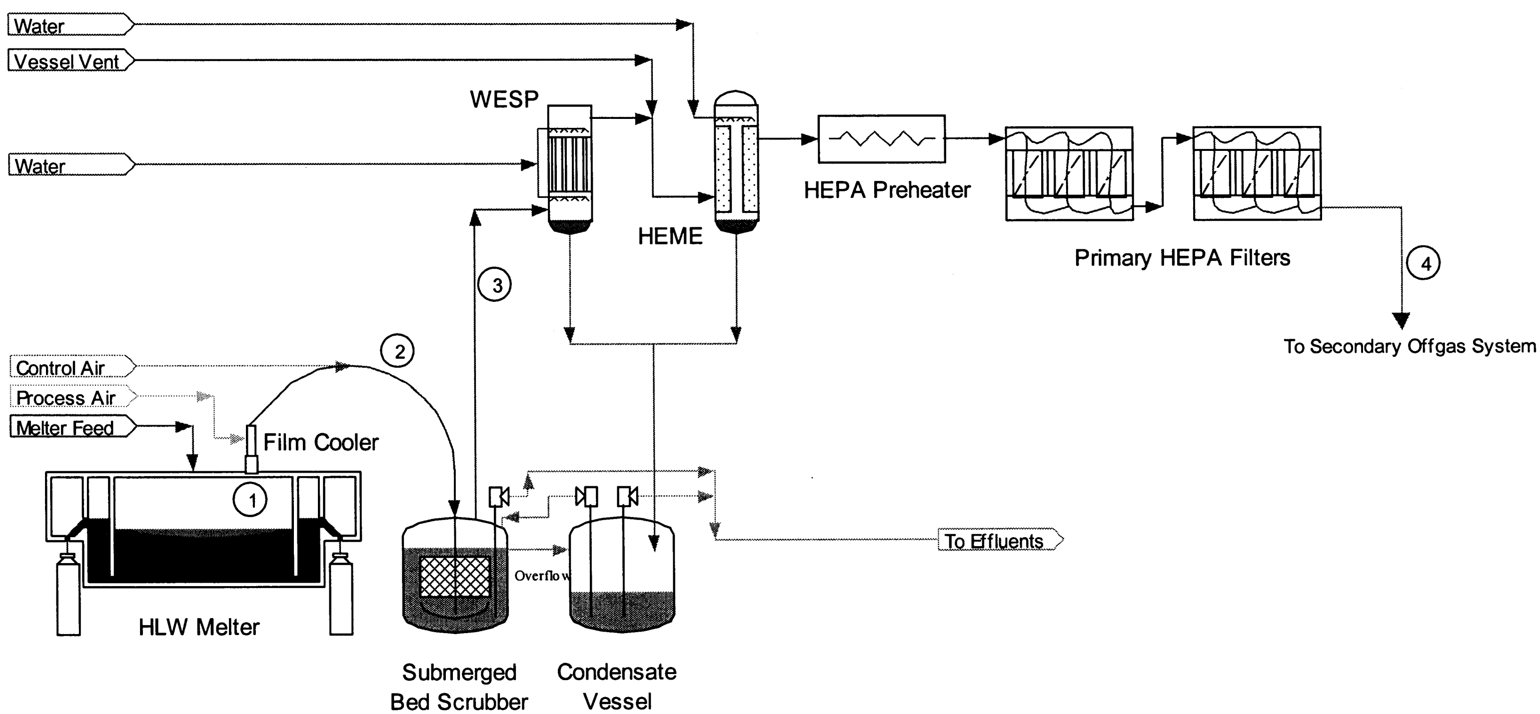

Figure 2. High-Level Waste Vitrification Primary Off Gas System

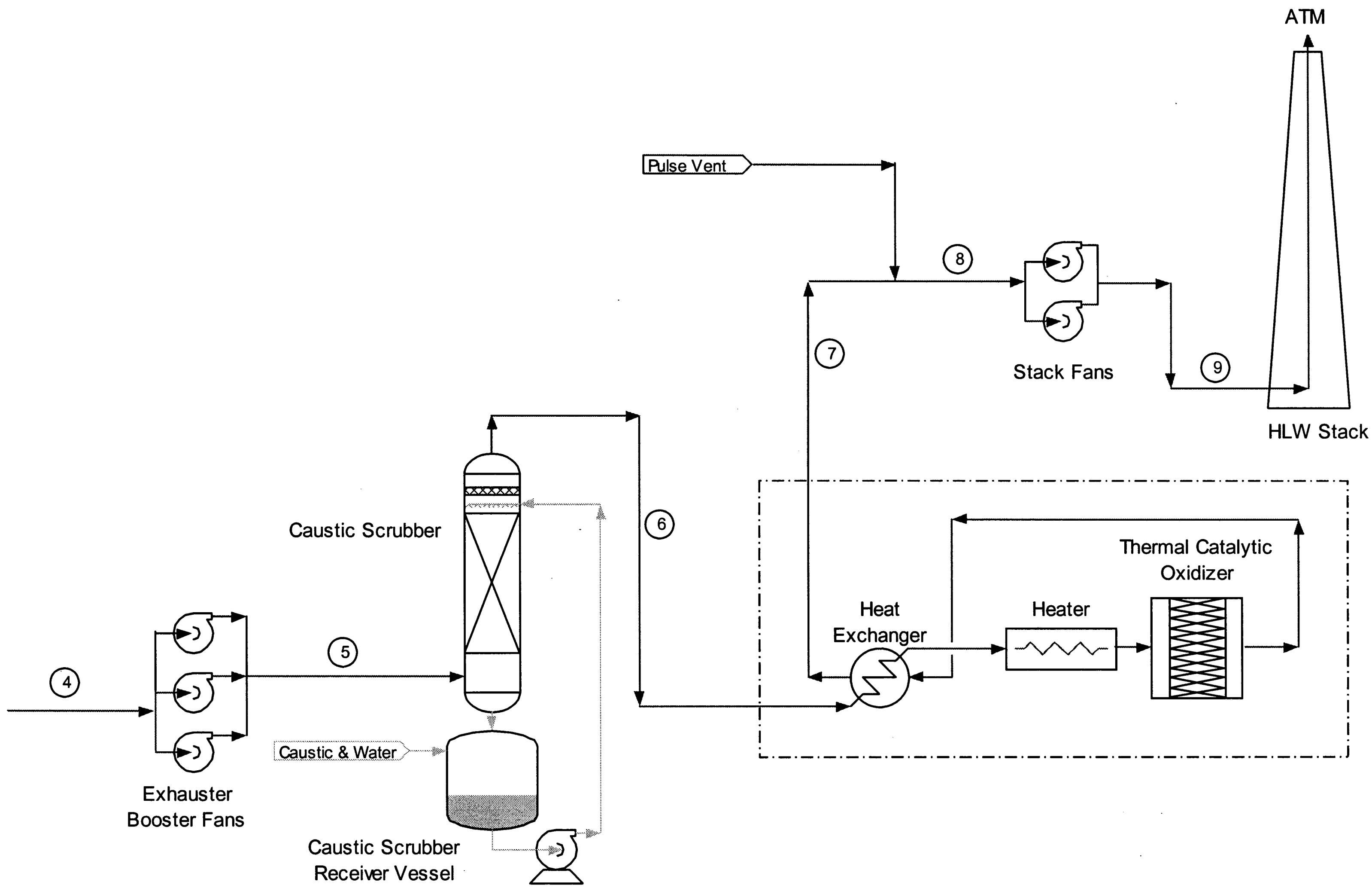

Figure 3. High-Level waste Vitrification Secondary Off Gas System

The bed-retaining walls extend below the support plate creating a lower skirt, to allow the formation of a gas bubble underneath the packing. The depth of the bubble is a reflection of the pressure drop across the inlet gas distribution plate and the packed bed. The entire bed is suspended off the floor of the SBS to allow the scrubbing solution to circulate freely through the bed. After formation of the gas bubble beneath the packing, the injected offgas then bubbles up through the packed bed. The rising gas bubbles also cause the scrubbing liquid to circulate up through the packed bed, resulting in a general recirculation of the scrubbing solution. The packing breaks larger bubbles into smaller ones to increase the gas/water contacting, thereby increasing particulate removal and heat transfer efficiencies. The warmed scrubbing solution then flows downward outside of the packed bed through coiling coils and past a cooling jacket which surround the bed. Because the offgas is cooled below its dew point, the scrubbing solution is supplemented by the resulting condensate.

The SBS offgas is routed to the Wet Electrostatic Precipitator (WESP). The WESP removes aerosols and particulates down to submicron size. The WESP houses vertical tubes, which act as positive electrodes. Each of these tubes also has a single negatively charged electrode, which runs down the centerline of each tube. A high voltage transformer supplies the power to these electrodes. To insure a uniform distribution of the inlet offgas into the electrode tubes, a gas distributor is used. The strong electromagnetic field generated by the tube electrode wire (or rod) applies a negative charge to the offgas particles. These negatively charged particles are then attracted to the positively charged tube walls. Collected particles are washed from the tube wall by condensate that is also collected on the inner tube wall. The quantity of "washing" condensate will be boosted by passing cooling water on the shell side of the electrode tubes, decreasing the saturated offgas temperature and causing condensation.

The purpose of the High Efficiency Mist Eliminators (HEMEs) is to further remove radioactive aerosols from the HLW melter offgas stream, the vessel ventilation system, and to reduce the dust-loading rate on the primary HEPA filters. A HEME is essentially a high efficiency demister that has a removal efficiency of approximately 99% for aerosols down to the sub-micron size (<0.1 μm). As the offgas passes through the HEME, the liquid droplets and other aerosols within the offgas interact with HEME filaments. As the aerosols contact the filaments they adhere to the filaments surface by surface tension. As the droplets agglomerate and grow, they eventually acquire enough mass to fall by gravity to the bottom of the unit, thus overriding the original surface tension, friction with the filaments, and the gas velocity. These collected droplets are assumed to contain the majority of the offgas's radioactivity and will be collected in the bottom of the HEME. As the condensate flows down through the filter bed, a washing action is generated that will help wash collected solids from the filter elements. However, some solids may accumulate in the bed over time, causing the pressure drop across the filter to increase. When the pressure drop across the HEME reaches a predefined level, it will be taken offline and washed with process water to facilitate removal of accumulated solids. Insoluble solids are assumed to remain, however, and their accumulation will eventually lead to the entire replacement of the HEME filter bed. A figure of a typical HEME is shown in Figure 5.

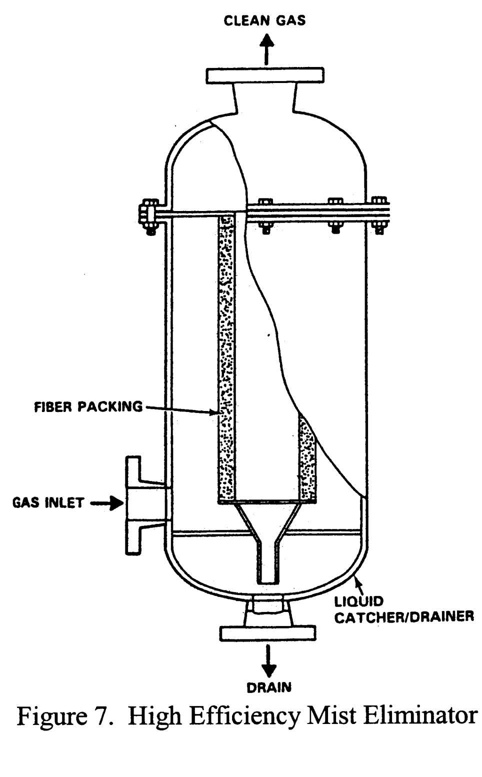

Figure 7. High Efficiency Mist Eliminator

Final submicron particulate removal is provided by a dual bank of high efficiency particulate air (HEPA) filters in series. This dual bank of HEPA filters is expected to have particulate removal efficiency in excess of 99.999%.

To remove residual volatile organic compounds (VOCs) and acid gases in the filtered HLW off gas stream, a packed bed caustic scrubber column and a thermal catalytic oxidizer unit are employed in the HLW secondary off gas system. The caustic scrubber will remove residual acid gases present on the melter off gas. The thermal oxidzier unit allows the oxidation of VOC compounds at a relatively low temperature of ~300 °C using a platinum metal catalyst. As with all thermal oxidation reactions, the organic compounds are converted to carbon dioxide and water vapor. The thermal oxidizer unit is composed of three components, a heat recovery exchanger, an electric heater, and the thermal catalyst bed. The electric heater is employed to heat the column feed to the desired oxidation temperature. To help conserve energy, the hot exhaust from the unit is used to preheat the feed in an energy recovery exchanger, reducing the electric heater's load. This also helps cool the column exhaust. It is expected that over time the catalyst bed will loss effectiveness and will require replacement. The catalyst bed will therefore be designed for the manual removal and replacement when required or after a predetermined life span.

Mass, Heat, and Pressure Balance Development

Mass, heat and pressure balance calculations were performed in an Excel spreadsheet to estimate material flowrates and physical properties through the HLW vitrification facility. All calculations performed in this spreadsheet are performed using fundamental chemical engineering principles and assumes ideal behavior for both gas and liquid phases. This spreadsheet tracks 102 chemical and 29 radionuclide species from HLW feed receipt from the Pretreatment facility through stack discharge. Over 50 separate process streams are defined in the HLW material balance spreadsheet. Information from this spreadsheet was the primary design criteria for sizing all off gas treatment equipment. For each process stream, the following fluid properties are defined:

- Mass Flowrate
- Density
- Volumetric Flowrate
- Individual Component Concentrations
- Pressure (Gas Streams Only)
- Temperature
- Relative Humidity (Gas Streams Only)
- pH (Liquid Streams Only)

Select process streams from the HLW off gas material balance are included in Table 1. Stream numbers correspond to numbers identified on Figures 2 and 3.

Table 1. Select HLW Off Gas Process Stream Definition [Rouse(b)]

Stream Number	Mass Flowrate (kg/hr)	Volumetric Flowrate (m^3/hr)	Pressure (mbar)	Temperature (°C)	Relative Humidity (%)
1	880	2,100	985	400	0.2%
2	2,000	3,100	977	220	1.2%
3	1,800	2,100	878	50	100%
4	2,100	2,500	861	69	33%
5	2,100	2,300	959	77	27%
6	2,100	2,000	952	44	100%
7	2,100	2,700	946	140	0.1%
8	5,900	6,100	943	61	17%
9	5,900	5,800	1009	66	14%

Aerosol Gas Collection Efficiencies

An integrated approach was used to determine aerosol collection efficiencies for the different HLW off gas treatment units as is suggested by Heumann. This method requires both a detailed understanding of the initial particle size distributions from the HLW melter and the collection efficiencies for each treatment, also as a function of particle size. By using this method, realistic collection efficiencies can be developed for each treatment unit taking into account the efficiency of the preceding treatment units. The fundamental equation used to calculate unit collection efficiencies is defined from Heumann and is defined as follows:

$$E_T = \int_0^{\infty} \frac{\partial g(D_P)}{\partial D_P} f(D_p)\, \partial D_P \qquad (1)$$

Where,

E_T	=	Treatment Unit Overall Collection Efficiency
D_P	=	Particle Size (μm)
$g(D_P)$	=	Inlet Particle Size Mass Distribution
$f(D_P)$	=	Treatment Unit Collection Efficiency

Using this process, process and mechanical considerations such as superficial velocity, contacting lengths, etc., can also be taken into account in determining aerosol collection efficiencies. To determine the discharge particle size distribution from each process unit, the following equation was developed from Equation 1. This equation is then used to predict the inlet particle size distribution for the next treatment unit.

$$g(D_P)_{P2} = \int_0^{D_P} \frac{\partial g(D_P)_{P1}}{\partial D_P} \frac{(1 - f(D_P))}{(1 - E_T)} \partial D_P \qquad (2)$$

Where,

P1	=	Inlet Particle Size Mass Distribution
P2	=	Discharge Particle Size Mass Distribution

The use of these equations requires the use of Math application type software such as MathCAD. However, alternatives to Equations 1 and 2 were also developed using summations which could be applied to a "spreadsheet" calculations. Collection efficiencies determined through spreadsheet calculations were determined to be roughly equivalent to a MathCAD output as long as the summation intervals exceeded 100.

Future Development Work

Prototypical HLW Off Gas System

To further validate the HLW melter off gas design, a prototypical off gas system has been designed and built as part of the DM-1200 pilot melter, which is located at the Vitreous States Laboratories, Catholic University, Washington, D.C. This melter and off gas system has been designed to be a third scale pilot melter for the HLW vitrification facility. The off gas system contains a SBS, WESP, HEME, HEPA filters, packed bed scrubber, and a thermal catalytic oxidizer skid. Commissioning and operation of the HLW pilot melter and off gas system will commence this fall.

Initial commissioning tests will primary focus on resolving design issues related to the submerged bed scrubber. Submerged bed scrubbers were initially developed and used at Pacific Northwest National Laboratory. They are currently in use at the West Valley vitrification facility and the Tokai-Mura nuclear reprocessing facility in Japan. They are passive devices, providing a relatively high degree of aerosol removal and off gas quenching. Throughout their development and operation, however, two fundamental concerns with the use of SBS technology have become apparent. The first is the pressure surging induced on melter pressure control systems. This is caused by the dual phase slug flow patterns through the SBS packed bed. Through the use of "liquid weirs" installed in the bottom gas distribution plate, it is hoped that both the amplitude and frequency of these surges can be minimized. The second is the potential for captured solids to accumulation in the bottom of the SBS, eventually leading to bed pluggage. Through the use of water jet nozzles placed near the bottom of the West Valley SBS, it has been shown that solids can be suspended and withdrawn through a suction device located at the very bottom, center, of the SBS vessel. A continuation of this concept has been installed in the DM-

1200 SBS. Offline testing of this solids removal system using a simulated sludge have shown that the system works effectively. Further tests will be performed to optimize the system when the DM-1200 melter is in operation.

Additional testing on the DM-1200 off gas system will focus on maintainability/operational issues, aerosol removal efficiencies, and solids accumulation throughout the off gas equipment and ducting. The validity of material balance calculations and assumptions will also be examined by applying the same methodologies to the DM-1200 melter and off gas system performance. Off gas analysis will also be performed to better characterize speciation of off gas contaminates, especially halogens such as iodine.

Dynamic Off Gas Pressure Model

A dynamic off gas pressure simulation model is being developed to simulate system response to changes in off gas flowrates. The simulation is being developed using Aspen Speedup and models both air and water vapor throughout the off gas system. Both gas flowrates and pressures are calculate using fundamental mass balance and fluid dynamic equations. The model can be used to develop pressure/flow response curves at any point within the system after a system upset such as a melter off gas surge. This model will be used to develop pressure control strategies and to optimize the design.

Conclusions

An effective and reliable melter off gas system has been developed for the RPP-WTP HLW vitrification facility. This system utilizes proven technologies both from the nuclear industry, as well as from the commercial arena. The primary off gas system is completely passive, while having a very high aerosol collection efficiency. This allows for the secondary off gas system to be contact maintained, thereby reducing operating expenses. Given this, optimization of the system design is still being performed through the resolution of outstanding issues. These include solids accumulation within the off gas ducting and submerged bed scrubber, pressure surging caused by the dual phase flow through the submerged bed scrubber, and contaminate speciation within the melter off gas. However, through research and development efforts and pilot plant operation, all of these issues can be resolved.

References

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