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Steam Reforming Application for Treatment of DOE Sodium-Bearing Tank Wastes at Idaho National Laboratory for Idaho Cleanup Project

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ABSTRACT

The patented THOR® steam reforming waste treatment technology has been selected by the U.S. Department of Energy (DOE) as the technology of choice for treatment of about one million gallons of sodium-bearing waste (SBW) at the Idaho National Laboratory (INL) Site [1]. SBW is an acidic waste created primarily from cleanup of the fuel reprocessing equipment at the Idaho Nuclear Technology and Engineering Center (INTEC) at the INL. SBW contains high concentrations of nitric acid and alkali and aluminum nitrates with minor amounts of many inorganic compounds including radionuclides, mainly cesium. The steam reforming process will convert the SBW into dry, solid, carbonate and aluminate minerals supporting a preferred path for disposal as remote handled transuranic (RH-TRU) waste at the Waste Isolation Pilot Project (WIPP).

The Idaho Cleanup Project (ICP) will design, build, and operate an Integrated Waste Treatment Unit (IWTU) that will comprise an integrated THOR® process system that will utilize dual fluidized bed steam reformers (FBSR) for treatment of the SBW. The IWTU is being constructed at INTEC, immediately east of the New Waste Calcine Facility (NWCF). Detailed design of the IWTU has been completed and DOE has approved the CD-3 detailed design. The State of Idaho has approved the RCRA and construction air permits. Construction of the IWTU started in April 2007 with civil and foundation work. This paper provides a project and process overview of the IWTU and discusses the design and construction status. IWTU equipment and facility designs and bases will be presented.

INTRODUCTION

In March of 2005, the DOE announced that CH2M-WG Idaho, LLC (CWI) had been selected to lead the $2.9 billion environmental cleanup of the INL Site. CWI is comprised of Denver-based CH2M HILL and Boise-based URS Washington Division (formerly Washington Group International [WG]). The cleanup project, named the Idaho Cleanup Project (ICP), is a seven-year undertaking to perform the cleanup of the 890-square-mile site. As the ICP contractor, CWI is responsible for treatment and disposal of radioactive waste; retrieval, disposal, and other remediation related to buried waste; safe management of spent nuclear fuel; disposition of nuclear materials; disposition of reactor and non-reactor nuclear facilities; and other environmental remediation activities. Included in the Scope of Work (SOW) for the ICP is the treatment of approximately one million gallons of SBW stored in three underground tanks.

[1] Certain information addressed within this paper pertains to Contract No. DE-AC07-05ID14516 between CH2M-WG Idaho, LLC and the U.S. Department of Energy. The views and opinions of the authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

The DOE issued the Record of Decision (ROD) selecting steam reforming as the treatment process for SBW in December 2005 [1]. In the ROD, the DOE stated that the preferred disposal path for the SBW would be as remote-handled transuranic (RH-TRU) waste, but that "Until such time as the regulatory approvals are obtained and a determination that the waste is TRU is made, the Department will manage the waste to allow disposal at WIPP or at a geologic repository for spent nuclear fuel (SNF) and HLW."

In order to meet the requirement to provide dual disposal paths for the SBW, CWI proposed to treat the SBW with the patented THOR® steam reforming waste treatment technology. The THOR® technology can produce a final waste product that is suitable for disposal as RH-TRU; and with modifications, it can produce a different waste form that could be qualified for ultimate disposal in a high-level (HLW) geologic repository. CWI selected THOR Treatment Technologies, LLC (TTT), an affiliate of URS Washington Division, to design and build a THOR® steam reforming waste treatment system for SBW (Figure 1 provides a drawing of the IWTU at the INTEC facility). The THOR® system will produce a waste form suitable for disposal as RH-TRU. The remainder of this paper provides an overview of the THOR® system design for that preferred disposal path. If the DOE calls for disposal of the SBW as HLW, the THOR® system design would undergo several modifications to accommodate a HLW disposal path.

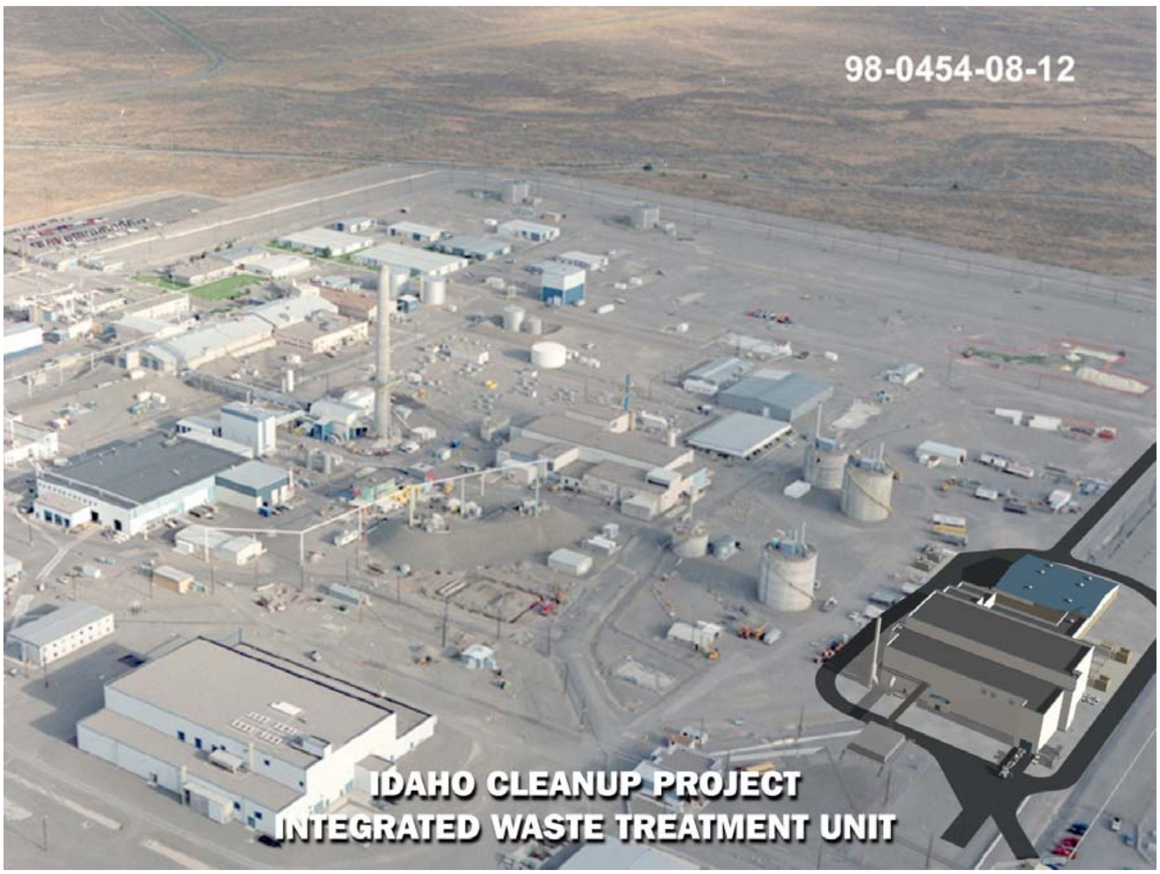

Fig. 1. Integrated Waste Treatment Unit housing the THOR® Steam Reforming Unit.

THOR® PROCESS OVERVIEW

SBW at INL consists of radioactive aqueous solutions with high concentrations of nitric acid, nitrates, alkali metals, aluminum, and a wide variety of other inorganic oxides. The THOR® steam reforming process destroys nitric acid, nitrates and organic materials present in the SBW, and produces a dry, solid product. The steam reforming process has been proven to efficiently handle, process, and immobilize the radionuclides, sodium, potassium, sulfate, chlorides, fluorides, and non-volatile heavy metals into a solid matrix [2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. The process converts nitric acid, nitrates, and nitrites directly to nitrogen gas in the steam reformers. Any organic material in the SBW is converted to carbon dioxide and water vapor in the steam reformers by a combination of steam reforming and oxidizing reactions. The THOR® process flow diagram for treatment of SBW is provided in Figure 2.

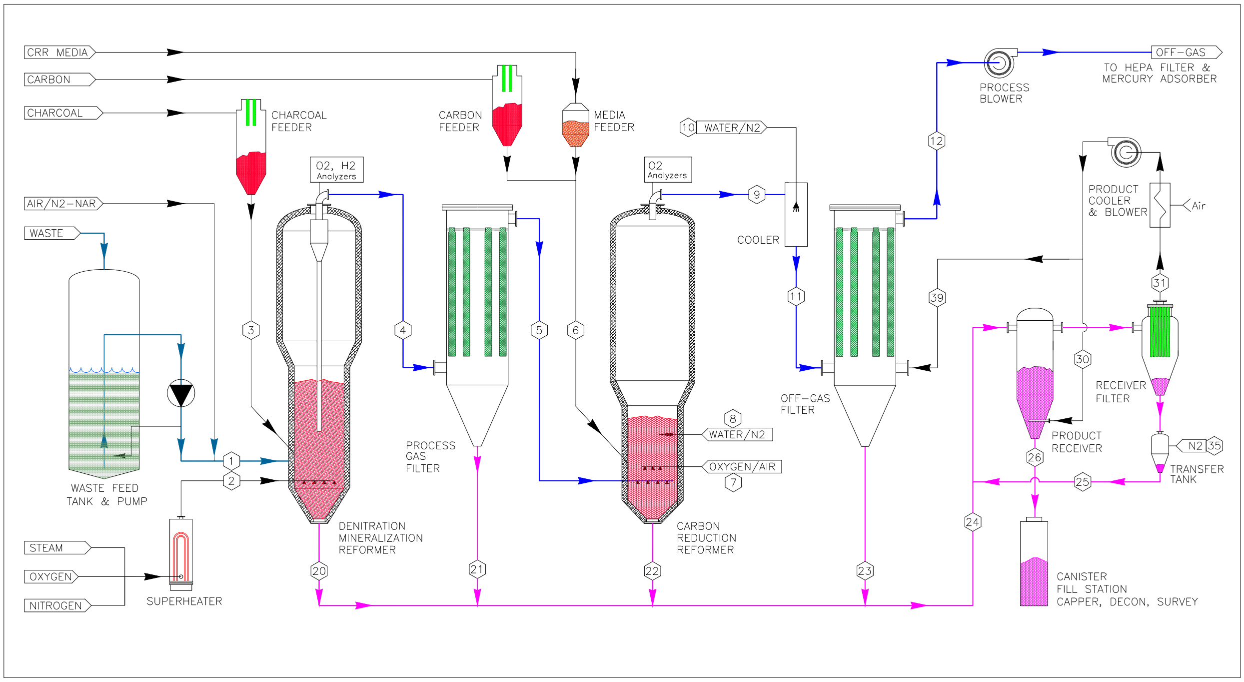

Fig. 2. THOR® process flow diagram.

The SBW tank-waste feed is introduced into the first steam reformer, the Denitration and Mineralization Reformer (DMR). The bed particles in the steam reformer are fluidized by introduction of near ambient pressure superheated steam. In the first steam reformer, liquids are evaporated; the vast majority of organics, nitric acid, nitrates, and nitrites are destroyed; and the reactive chemicals in the waste feed are converted to a solid waste product. The second reformer, the Carbon Reduction Reformer (CRR), serves to further reduce any NOx gases from the first reformer and oxidize residual organics (if present) to carbon dioxide and water vapor. The gases (mainly carbon dioxide and water vapor) from the process are filtered through high-efficiency sintered metal filters, HEPA filters, and a mercury adsorber. They are then vented to the atmosphere through a monitored stack. The THOR® final end-product, an alkali carbonate and aluminate solid, will meet requirements for shipment in the RH-72B shipping container. It is expected that the THOR® end-product will meet anticipated requirements for disposal as RH-TRU.

For the ICP SBW processing effort, the alkali metals present in the SBW will form mainly carbonates and aluminates and the inorganic constituents will form metal oxides and spinels with the iron in the SBW. The carbonates are made from the adsorption of carbon dioxide (from the steam reforming reactions in the DMR), and aluminates are formed with the alkali metals and aluminum, respectively, from the SBW. The combined SBW inorganic minerals are largely carbonates and will have melting points in the 1,023°K to 1,123°K (750°C to 850°C) range. Therefore, in order to reliably convert SBW constituents into carbonates, the DMR will be operated at no higher than 953°K (680°C) to prevent the carbonates and other salts from partially melting or becoming sticky in the DMR.

PROCESS EQUIPMENT DESCRIPTION

Denitration and Mineralization Reformer

The DMR is operated at near ambient pressure and will maintain an average bed temperature of approximately 913°K (640°C) when producing a carbonate-rich product. Superheated steam, enriched with oxygen, is injected into the bottom of the DMR. The low-pressure steam fluidizes the granular carbonate product particle bed materials in the vessel by means of pipe distributors located above the bottom head of the vessel.

The waste is fed into the DMR through injection ports just above the fluidizing gas (oxygen enriched steam) distributor. The waste feed is atomized into the vessel, and the water in the waste feed is instantly evaporated and superheated to the bed temperature by the large mass of hot, fluidized product solids. The resulting dried waste solids quickly heat to reforming temperatures. Any organics in the feed are volatized and pyrolyzed upon contact with the hot bed solids. A cyclone gas/solid separation device is installed in the top of the DMR. This device allows the process gases to flow from the DMR to the Process Gas Filter (PGF) and then to the CRR and sends larger solids back to the fluidized bed portion of the vessel. The DMR is operated under reducing conditions throughout most of the bed and the entire freeboard zone. Granular carbon is fed to the DMR to provide the nitrate reducing process conditions and to provide the energy required to maintain the DMR at the desired operating temperatures. One nozzle is provided for addition of solid carbon and additives. Accumulated product granules are removed from the bottom of the DMR and are pneumatically transferred to the product receivers where solids are packaged into RH-72B canisters. The DMR is constructed of qualified high-temperature alloy metals.

The DMR, PGF, CRR, and related process equipment, shown in Figure 3, are located within two concrete shielded structures.

SBW Process Gas Filter

The process gas from the DMR flows to the PGF, which captures any carbonate-rich DMR product fines carried over in the process gas. The carbonate fines could cause agglomeration in the CRR as the melting point of the carbonate-rich SBW mineral product have a melting point range of 1,023°K to 1,123°K (750°C to 850°C). The filter will operate at 323°K to 373°K (50°C to 100°C) below the DMR temperature, and is constructed of qualified high-temperature alloy metals. The process filter elements are a sintered metal type that will efficiently remove particles from the DMR process gas. The filtered solids will be pneumatically transferred to the product receivers where the fines are combined with the granular solids removed from the bottom of the DMR.

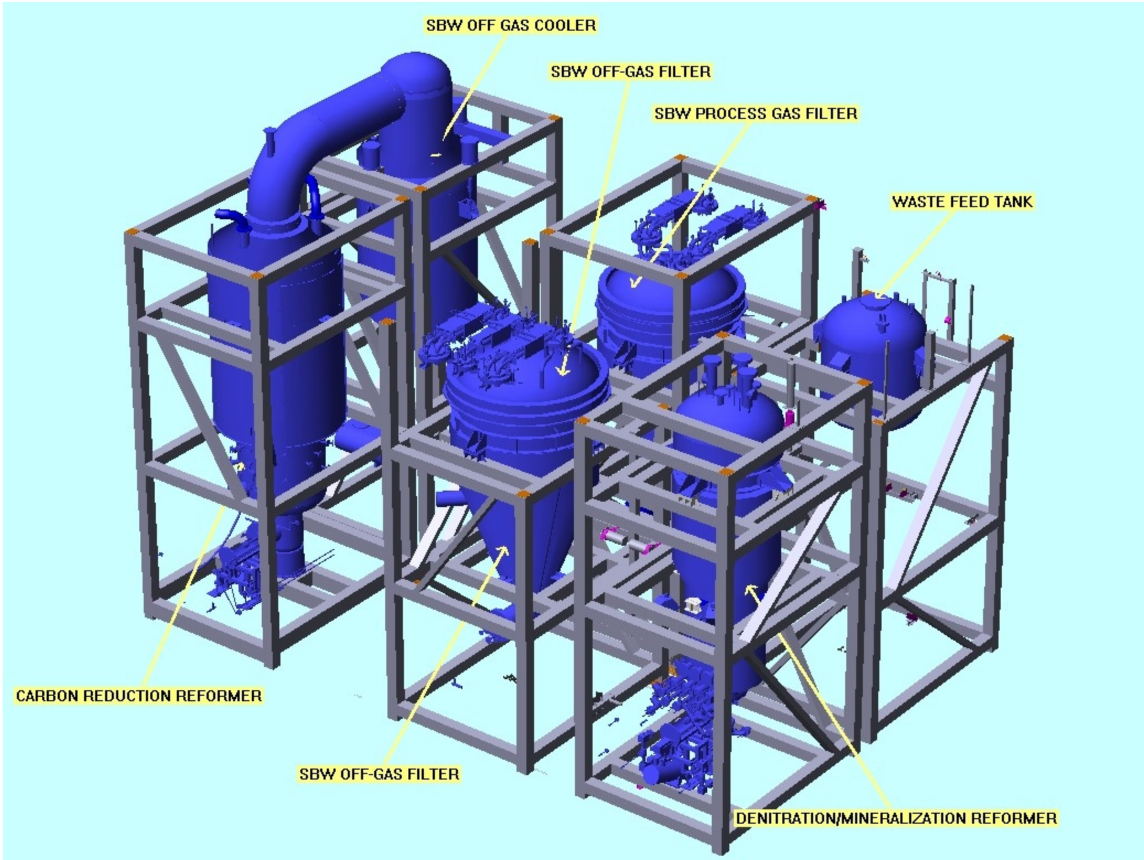

Fig. 3. Steam reforming process equipment module arrangement – shielding removed for clarity.

Carbon Reduction Reformer

The process gases from the DMR flow through the process gas filter to the fluid gas inlet distributors of the CRR located near the bottom of the CRR vessel. Auto-thermal gas (oxygen) is fed into the CRR bed. The CRR operates under both reducing and oxidizing conditions. A unique aspect of the CRR is that the bottom of the fluidized bed media produces a strongly reducing environment where residual NOx from the DMR is further converted to nitrogen gas by the presence of the reducing gases in the process gas stream used to fluidize the bottom of the CRR and addition of carbon to the CRR bed. Oxygen is injected into the upper portion of the bed to produce an oxidizing region where the organic materials are destroyed, and the hydrogen and carbon monoxide produced in the DMR are converted to carbon dioxide and water with very high efficiency. Because the CRR has no open-flame combustion, the temperatures throughout the fluidized bed are uniform and only minimal levels of thermal NOx are generated. The DMR and CRR reformers serve to essentially eliminate NOx emissions from the process with >95% efficiency.

The CRR bed media comprises an alumina granular media that forms a semi-permanent fluidized bed. Because essentially no solids are input with the DMR process gases into the CRR, the only increase in bed solids is from planned additions of media and carbon from the additive feeder system. The media is added only to make-up for bed media particle attrition and carryover (elutriation) of bed media particles to the off-gas treatment system. It is expected that no CRR bed material will be drained during normal operation for the duration of the SBW treatment campaign. A solids drain is provided on the bottom of the CRR to provide drain capability in the event maintenance must be performed on the fluidizing or oxygen gas distributor located in the bottom of the CRR.

Under normal operating conditions, the CRR temperature is 1,173 to 1,273°K (900 to 1,000°C) and operates under vacuum at the top of the bed. Off-gas from the CRR enters the off-gas treatment system where a water spray cooler quickly cools the hot off-gas to 423°K to 473°K (150°C to 200°C). All of the water is evaporated and is carried with the cooled off-gas as water vapor to the Off-Gas System.

OFF-GAS SYSTEM

Process Vent High-Efficiency Particulate Air Filters

The process off-gas is first filtered through the Off-Gas Filter (OGF) and then through the high-efficiency particulate air (HEPA) filters. Filtered gases then flow to the mercury adsorber.

Mercury Adsorber

The Mercury Adsorber is a dual bed granular activated carbon (GAC) unit designed to remove mercury present in the off-gas. Dual beds are provided so that spent GAC media can be replaced with new media when the first bed adsorbent reaches capacity. The spent carbon bed media will be disposed of in accordance with regulations and a new charge of carbon media will be loaded as required to maintain removal efficiency.

Continuous Emission Monitoring System

The off-gas from the process will be monitored prior to discharge. Off-gas samples will be continuously taken and the off-gas sample stream will be analyzed in the Continuous Emission Monitoring System (CEMS) as required by the air permit. The off-gas is then combined with the filtered air from the building ventilation HEPA filter system and the combined off-gas and ventilation air stream will be discharged through the stack. Radiological and NOx monitors will be provided on the stack. Based on documented performance of the THOR® process, the off-gas from the THOR® steam reforming process will be compliant with Maximum Achievable Control Technology (MACT) standards and State of Idaho air permit limits.

Building Ventilation System

The process cells and building ventilation flows are filtered by the Building HEPA Filters. To change filter elements, one of the banks of filters is isolated and the filter elements changed out. The discharge from the Ventilation Blowers is vented to the atmosphere through the monitored stack. The discharge air from the Ventilation Blowers is mixed with the process off-gases and then discharged to the stack where the gases are monitored with a radiation and NOx monitors.

SBW Product Receiver/Cooler

The two Product Receiver Cooler (PRC) vessels receive granular and fine particulate solids from the SBW treatment systems. Cool nitrogen passes through the PRCs to cool the product. The temperature of the product is monitored. Product solids accumulate in the bottom of the PRCs. The accumulated solids may be sampled and then transferred by gravity to the RH-72B canister. The vented nitrogen from the PRCs flows to the Off-gas Filter and then to the off-gas system for final filtration by the HEPA filters. The SBW waste carbonate PRCs and RH-72B canister loading cells are shown in Figure 4.

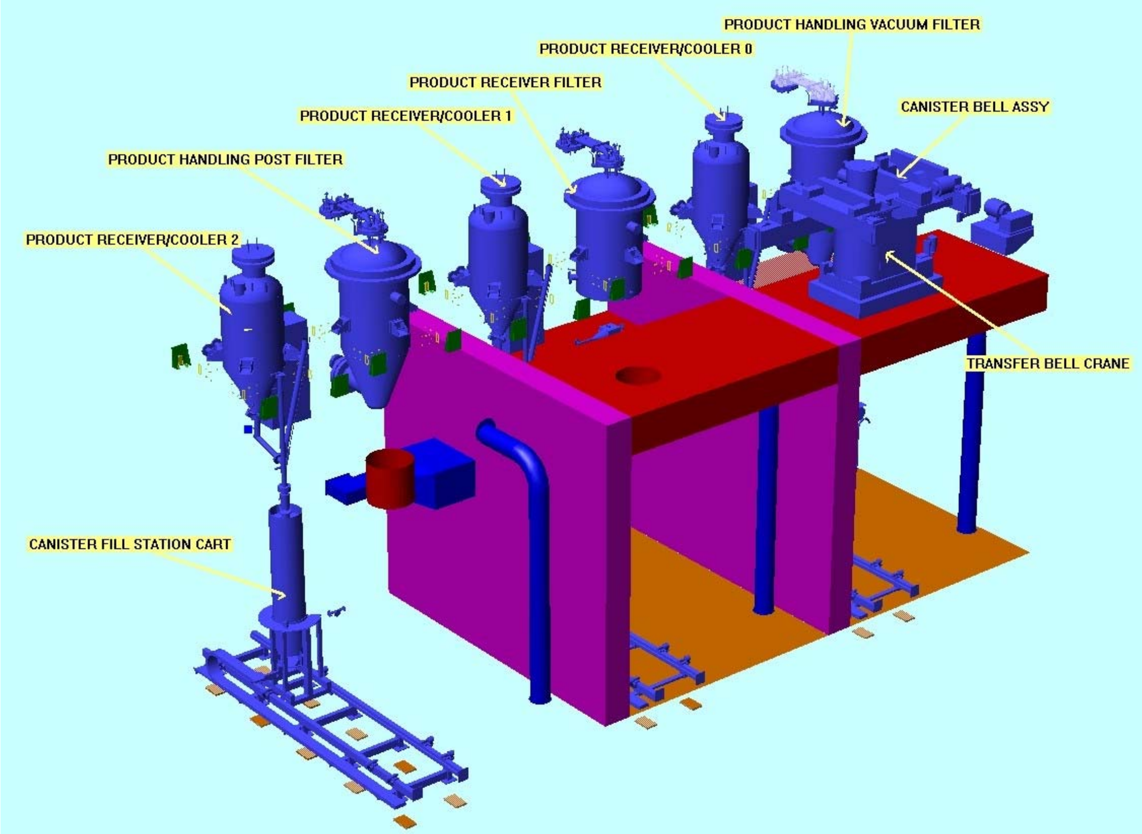

Fig. 4. SBW Product Receivers and Canister Loading Cells.

Canister Fill Station

The Canister Fill Station consists of two parallel trains of equipment to fill, cap, and decontaminate canisters for storage of the processed waste. Each of the two parallel trains consists of the following: a removable shield plug to receive canisters into the cell, a canister fill station transfer cart to move the canisters through the filling and capping process, a canister sealing chute to prevent leakage to the outside of the canister, a canister capping station to close/seal the canister, and a canister swipe and decontamination system to clean any potential product contamination off the canister. All operations are remote and in-cell.

Canister Transfer and Storage

Filled and surveyed canisters are transferred from the fill station cells to concrete storage vaults by means of a heavily shielded, rail mounted, transfer bell and carrier crane. Each vault holds 16 canisters. Once the storage vault is loaded with 16 canisters, the concrete lid is lowered into position, and the loaded and closed storage vault is transferred to the separate, interim storage building, located adjacent to the process facility on the north side. When the canisters are to be shipped, the storage vault is returned to the process building and the transfer bell transfers each canister into an awaiting RH-72B cask. The loaded cask is closed, surveyed, and released from the IWTU for shipment.

STATUS OF IWTU PROJECT

The IWTU Project has achieved a number of significant milestones and is proceeding on plan for meeting the project milestones. The DOE has approved Critical Decision (CD) - 1, the associated conceptual design, the CD-2, Performance Baseline, preliminary design package, and the CD-3, Detailed Design. Construction of the IWTU commenced in April 2007. Figure 5 shows a perspective view of the IWTU Facility.

Fig. 5. Integrated Waste Treatment Unit Facility.

Procurement activities are underway with construction of the long-lead pressure vessel having commenced in early 2007. The RCRA and air permits have been received from the State of Idaho.

An extensive series of pilot scale demonstrations of the THOR® steam reforming process utilizing the entire thermal process flow sheet have been completed and results have been published and lessons learned incorporated into the IWTU detailed design. [12, 13, 14]

The IWTU Project schedule is based on 26 months from contract signing, May 2005, to submittal of the CD-3 for construction approval and 30 months from CD-3 submittal to CD-4, Approval for Hot Operations, with a DOE contingency of an additional 7 months.

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