

Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

SRNL-L3300-2019-00027

July 11, 2019

To: J. G. Reynolds

From: M. E. Stone

Technical Reviewer:

R. B. Wyrwas

Date

Approval:

J. Manna

Date

Review of Submerged Bed Scrubber Assumptions in the Hanford Waste Treatment and Immobilization Plant Bases and Requirements Document

A review was performed of the assumptions in the Hanford Waste Treatment and Immobilization Plant Bases and Requirements Document (BARD) for the Low Activity Waste (LAW) Submerged Bed Scrubber (SBS). This review was performed to evaluate how the assumptions impact the TOPSim process models that utilize the assumptions for system modeling.

The RPP Integrated Flowsheet uses TOPSim¹ to model the entire waste retrieval and treatment process for the Hanford tank waste. This model relies on a number of inputs, including the BARD². The BARD represents the basis for the WTP process models; as such, it contains some assumptions that are valid for the models that will not accurately reflect actual operation. For example, the amount of LAW condensate generated is overstated in the BARD to allow for conservatism in the design of the condensate handling unit operations. The assumptions in the BARD for the LAW SBS were reviewed to evaluate how actual operation could differ from the design basis assumptions.

It should be noted that this review did not evaluate nor is it intended to address issues related to design of the WTP facility such that the information in this report is should not be viewed as issues with the design of the WTP facility nor should the information be used without additional evaluation to generate design concerns.

We put science to work.™

Summary

The review did not identify any areas of concern for the way the BARD assumptions for the SBS are used during process modeling. It was noted that the operating conditions during pilot plant testing were above the scaled values expected during full-scale operation for non-condensable flow through the SBS. In addition, condensable flowrates may be higher than tested in the pilot scale tests due to the more dilute feed planned for DFLAW operations. The lower non-condensable flow for full scale operations and the higher condensable flowrate would be expected to improve particulate DF. pH and temperature were consistent during pilot plant testing and this review did not identify any concerns that the full-scale process would differ significantly from the pH ranges noted in pilot scale data.

It was noted that the assumed Decontamination Factors (DFs) for components in the offgas stream should not be considered bounding and are the best available estimates for the expected overall average system DFs based on pilot plant data. A number of species do not have non-radioactive isotopes and rely on identified surrogates (for example, non-volatile radioactive species are assigned a DF based on the average DFs of Al, Fe, and Zn). In addition, the DF values were determined for normal operations of the melter and did not differentiate between periods of typical operation and idle periods. The assumed DFs will be validated during commissioning of the LAW facility.

Two previously identified gaps were not closed by this review: 1) Iodine speciation and partitioning in the melter and offgas system and 2) mercury partitioning in the offgas system.

We put science to work.™

SBS Unit Operations Background

The LAW SBS is utilized to condense water and scrub particulate from the LAW melter offgas³. The SBS receives the melter offgas directly from the melter film cooler and passes the offgas through a packed column submerged in offgas condensate, as shown in Figure 1. The offgas is cooled to approximately 50 degrees Celsius in the scrubber, condensing most of the water. Particulate is scrubbed from the offgas by a combination of the condensation of the water from the offgas and contact of the offgas with the condensate in the packed columns. Cooling coils are used to control the SBS temperature to approximately 50 degrees Celsius. Collected condensate overflows from the SBS to a condensate vessel. Condensate in the collection tank is recirculated from the SBS vessel to aid in cooling and provide mixing in the SBS.

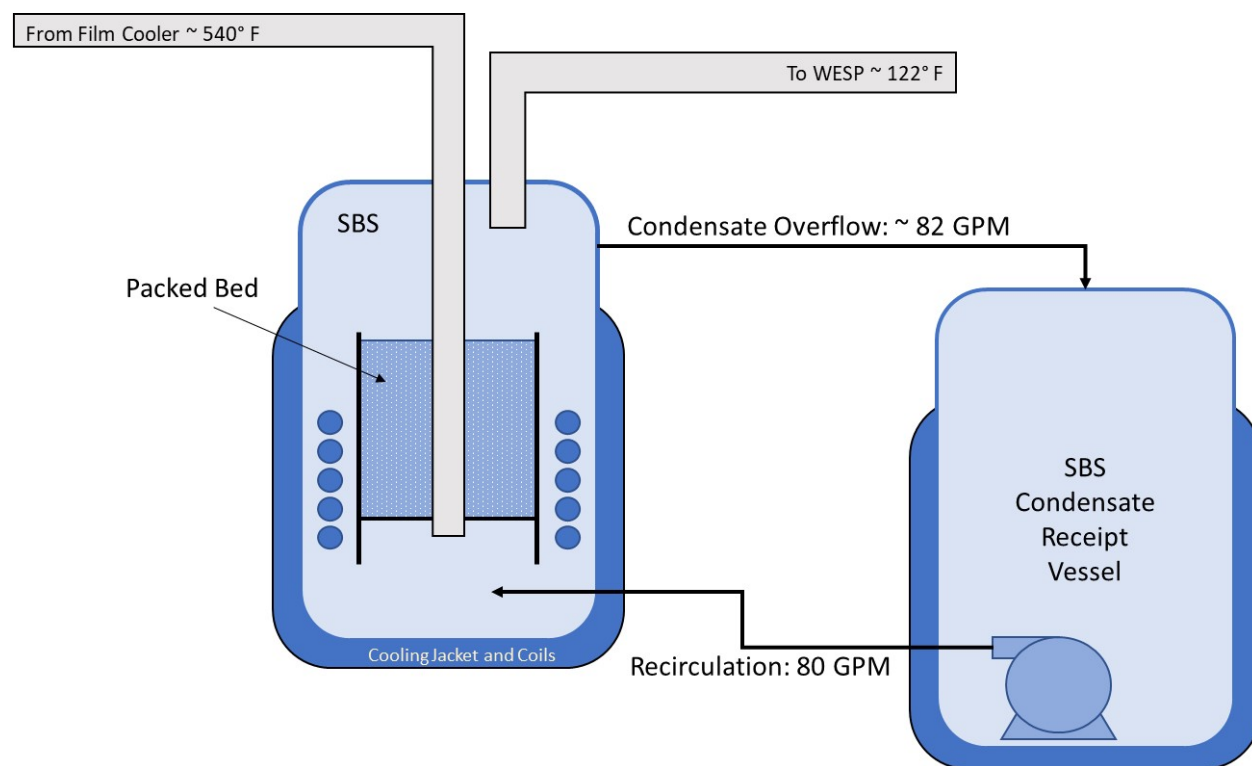


Figure 1. Simplified SBS Diagram

We put science to work.™

Discussion

Water Removal

The BARD assumes that each SBS will condense two gallons of water per minute from the melter offgas. The melter feed water content and melter feed rate determine the amount of water that must be condensed. The relatively dilute feed (nominally 5.6M Na) during DFLAW operations as well as the increases in waste loading expected from advanced glass models will result in feed that is more dilute than would be expected during full WTP operations.

Assuming feed at 5M Na, 30% soda loading in glass, a melter feed density of 1.25 g/ml, and 15 MT/day of glass per melter, the SBS condensate load would be approximately 4 GPM. However, it is not likely that the melter would be capable of maintaining design basis throughput with dilute feed at high waste loadings and the production rate limitations of the melter will result in practical limitations on amount of water sent to the SBS.

It is also noted that TOPSim modelling performed for system evaluations do not use the BARD assumption, but calculate the water content from the melter feed and feed rates. The models include limits for the melt rate of dilute feed as well as the cooling capacity of the SBS¹.

Decontamination Factors

Particulate Matter

As noted above, the one of the primary purposes of the SBS is to remove particulate matter from the melter offgas. In addition to understanding if the Decontamination Factor (DF) values are nominal or bounding, it is important to understand how the operational assumptions for the SBS may differ from the BARD assumptions and how those differences would impact the assumed DFs.

BNI identified three primary factors that impact the particulate removal performance of the SBS⁴:

1. Particulate size
2. Non-condensable flowrate
3. Condensation rate

Particulate size is directly related to scrubbing efficiency with larger particles have a higher DF than smaller particles. The non-condensable flowrate impacts the residence time and gas velocity in the scrubbing sections and is inversely related to DF. Condensation is a primary means to capture particulate and is directly related to DF with high condensation rates leading to better efficiency.

Gaseous Species

The SBS scrubber will scrub some of the non-condensable gases emitted from the melter (HI, I₂, NO_x, CO, CO₂, SO_x, ammonia, etc.) The primary factors impacting scrubbing of gaseous species are listed below:

1. Condensate pH
2. Condensate temperature (if not maintained at a constant 50 degrees Celsius)

We put science to work.™

3. Non-condensable flowrate

Combining the list of factors for particulate and gaseous species scrubbing efficiencies leads to the following list of factors that will be reviewed for impacts on the SBS operation during actual processing conditions versus the assumptions in the BARD:

1. Particulate size
2. Non-condensable flowrate
3. Condensation rate
4. Condensate pH
5. Condensate temperature

Evaluation of Operational and SBS Feed Parameters

Particulate Particle Size

Section 3.2.4 of the BARD concluded that melter offgas emissions were not significantly impacted by melter scale and that it is “reasonable” to assume that the pilot scale data can be extrapolated to the full scale melter. Thus, it is assumed that the particle sizes during LAW offgas operation will be similar to pilot scale testing.

Non-Condensable Flowrate

The non-condensable flowrate consists primarily of air added to the melter and offgas system, but also consists of non-condensable gases (e.g. CO₂) generated during melter cold cap reactions. The nominal flowsheet calculations estimate this noncondensable flowrate from cold cap reactions to be ~5.5 scfm, an amount too small to impact residence time in the SBS compared to the air addition flowrates.

The LAW melter is maintained under a slight vacuum (~5 INWC) relative to the surrounding process room. Some air inleakage to the melter is expected during normal operations and is accounted for in the melter vacuum control protocols. Air is added to the film cooler to provide initial cooling of the melter offgas and air is added to control the melter vacuum. In addition, air is bubbled into the melter pool to increase mixing in the melter as well as air added to the melter feed pump purge. It should be noted that higher air inleakage rates would result in less melter control air additions. The expected amounts of each of the air additions has been estimated for the LAW melters, as shown in Table 1⁵.

Table 1. Estimated Offgas Flowrates

Description	Minimum Addition (SCFM)	Maximum Addition (SCFM)

We put science to work.™

Melt Pool Bubblers and ADS Pump Purge	19	19
Melter Air Inleakage	150	300
Melter Film Cooler Air Additions	185	185
Melter Vacuum Control Air	200	300
Total Air Additions to Melter	550	~800

As noted previously, the SBS DF data in the BARD is based on pilot plant data. Little information on melter air inleakage and other rates was noted in the pilot scale reports. For example, the DM1200 test report for envelope A testing⁶ notes that the melter pressure control air system was not fully functional for the testing and that total offgas flows were 100-250 scfm, with 10-80 scfm of water. The DM1200 is a ~1/8 scale system compared to the LAW melter, offgas flows would be expected to be approximately 70 to 100 scfm to match the flows shown above for full-scale operation. Similar statements on offgas flow are made in the reports for other DM1200 testing. It is noted that the values for full-scale operation are design estimates and may not reflect actual values for operation. The lower non-condensable flows expected for WTP LAW melter operations would aid in particulate matter and gaseous species removal from the offgas system.

Condensation Rate

As noted above, the condensation rate during DFLAW operation could be higher than the amounts assumed in the BARD due to the more dilute feed. Higher condensation rates in the SBS would aid in particulate matter removal from the offgas stream.

Condensate pH

The SBS condensate is expected to have a relatively neutral pH based on pilot plant data, but it is acknowledged that pH of the condensate can be impacted by melter operations. For example, the waste feed contains large amounts of nitrate and nitrite. Sugar is added to the feed to prevent excessive foaming in the melter from oxidized feed and much of the nitrate and nitrite react with the sugar to form nitrogen and ammonia. The sugar addition is not stoichiometric; some nitrate and nitrite will form NO_x which is emitted from the melter. If the sugar additions are lower than needed, excessive NO_x and reduced ammonia formation in the melter can lead to highly acidic condensate as seen during the recent PNNL lab-scale melter test with actual tank waste.

Assuming the melter system is operated in a similar manner to the pilot scale tests, the pH is expected to be in the same range as the pilot scale data.

We put science to work.™

Condensate Temperature

The condensate is maintained at a set temperature by cooling coils in the SBS and a heat exchanger on the SBS condensate recycle. It is expected that the full-scale system will be able to maintain the condensate at the desired setpoints despite the increased condensation load expected from the dilute feed from DFLAW, as described above.

Overall Operational Condition Summary

SBS operations could be different than assumed in the BARD for DFLAW operations. The condensable loading on each SBS will be higher than the nominal 2 GPM assumed in the BARD. In addition, the non-condensable gas flow rates are likely lower than the scaled flows utilized during pilot scale testing. These two factors could impact the assumed DFs in the SBS, but it is difficult to determine the overall impact. Overall, the operational differences should aid in removal of species from the melter offgas.

Decontamination Factor Review

The BARD lists Decontamination Factors (DFs) for each species for the SBS. These DFs are utilized during process modeling to determine the partitioning of species for the SBS operation. The assumed DFs are average values based on pilot scale data with a single value utilized for each species⁷. It is noted that the DFs achieved during pilot scale tests varied considerably, but it was not possible to develop correlations to account for the variations noted. In addition, many radioactive species did not have a non-radioactive isotopes that could be evaluated during the pilot scale tests and DFs for these species are estimated. For example, non-volatile species are assigned a DF based on the average DFs of Al, Fe, and Zn.

For most species, the DF in the SBS is not a major concern as no processing limits are associated with the capture of these species by the SBS. Removal of certain species is credited by the air permits and/or required to achieve other objectives such as capture of Tc in glass. It is noted that the performance of the SBS in species removal will be determined during commissioning to validate the design assumptions. As noted above, higher DFs could be expected based on the differences in condensate rate and non-condensable flowrates.

Mercury represents a special case for the SBS scrubber. First, no testing has been performed for mercury with a SBS. The DFs in the BARD for each mercury species are based on data from small scale tests with a venturi scrubber⁸. Thus, partitioning of mercury in the LAW offgas system has been designated as a gap by the Integrated Flowsheet⁹. However, the reason the Hg represents a special case is that the only significant purge point for mercury in the LAW system is the LAW offgas. That is, nearly all (96%) of the mercury fed to the LAW melter is assumed to be removed by the Hg passing through the SBS with eventual collection on the activated carbon beds downstream of the HEPA filters. Thus, increased capture

We put science to work.™

of Hg by the SBS would reduce the single pass purge rate for Hg and lead to increased Hg concentrations throughout the LAW melter feed, LAW melter, and EMF systems.

Iodine removal is also noted as a gap by the Integrated Flowsheet. The pilot scale tests were performed using potassium iodide in the melter feeds; actual speciation of iodine in the feed is uncertain. HI and I₂ have been noted in the LAW melter offgas during pilot scale testing; these species are expected to be removed with different efficiency in the SBS. Speciation of the iodine in the offgas stream during actual operation is uncertain.

SBS Chemistry Assumptions

The BARD lists a number of chemical reactions assumed to occur in the SBS; these reactions are typically acid/base reactions, gas absorption reactions, and solid dissolution reactions. The extent of the reactions are set by the DF for each species for simple gas absorption or dissolution reactions. Reactions of nitrogen species in the SBS are complex since multiple reaction paths are available depending on process conditions. The amount of ammonia, nitrite, and nitrate in the SBS feed will vary depending on melter processing conditions. Ammonia will exhibit some degree of deprotonation to ammonium and the split between nitrite and nitrate being variable depending on the pH of the SBS scrub solution as well as process temperature and other operational factors. The ammonia concentration in the SBS condensate, along with boron species, help to buffer the pH of the SBS condensate to a near neutral pH despite the absorption of a number of acid gases such as HCl, HF, and NO_x.

The BARD describes the calculations used to determine the amount of each nitrogen species in the offgas condensate as well as how to determine the pH of the resulting condensate. As with the assumed DF's, the predicted values for the nitrogen speciation will be confirmed during cold commissioning and should be assumed to be expected average values as predicted using the BARD assumptions rather than bounding values.

We put science to work.™

References

1. A.M. Schubick, J.K. Bernards, N.M. Kirch, S.D. Reaksecker, E.B. West, L.M. Bergmann, and S.N. Tilanus, "Topsim V2.1 Model Requirements, RPP-RPT-59470, Rev 1.," Washington River Protection Solutions, Richland, Washington, 2016.
2. Y. Deng, B. Slettene, R. Fundak, R.C. Chen, M.R. Gross, R. Gimpel, and K. Jun, "Flowsheets Bases, Assumptions, and Requirements," Bechtel National, Inc. River Protection Project Waste Treatment Plant, Richland, Washington, 24590-WTP-RPT-PT-02-005, Rev 8, 2016.
3. "LAW Primary Offgas (LOP) and Secondary Offgas/Vessel Vent (LVP) System Design Description," 24590-LAW-3ZD-LOP-00001, Rev 3, Bechtel National Incorporated, River Protection Project, Waste Treatment Plant, Richland, Washington, 2018.
4. W. Lenzke, "Decontamination Factor and Release Fraction Definition for RPP-WTP Offgas Treatment Equipment, 24590-WTP-RPT-ENV-01-014, Rev 0," River Protection Project, Waste Treatment Plant, Bechtel National Incorporated, Richland, WA, 2002.
5. N.E. Wilkins and A. Coulam, "LAW Melter Offgas System Design Basis Flowsheets," Bechtel National Inc. Waste Treatment and Immobilization Plant, Richland, Washington, 24590-LAW-M4C-LOP-00001, Rev 5, 2018.
6. K.S. Matlack, W. Gong, T. Bardakci, N. D'Angelo, and I.L. Pegg, "Integrated Off-Gas System Tests on the Dm1200 Melter with RPP-WTP LAW Sub-Envelope A1 Simulants," Vitreous State Laboratory, The Catholic University of America, Washington, D.C., VSL-02R8800-2, 2002.
7. R.O. Lokken, "Reconcile Hlw and LAW Secondary Offgas (Sbs and Wesp) Decontamination Factors,," AEM Consulting, Richland, Washington, CCN-153212, 2008.
8. R.W. Goles, W.C. Buchmiller, L.K. Jagoda, B.D. MacIsaac, and S. Rassat, "Test Summary: WTP Flowsheet Evaluation of Mercury-Containing Hanford Waste Simulant," Battelle - Pacific Northwest Division, Richland, Washington, WTP-RPT-122, Rev 0, 2004.
9. L.W. Cree, J.M. Colby, M.S. Fountain, D.W. Nelson, V.C. Nguyen, K.A. Anderson, M.D. Britton, S. Paudel, and M.E. Stone, "One System River Protection Project Integrated Flowsheet," Washington River Protection Solutions (WRPS) One System, Richland, Washington, RPP-RPT-57991, Rev 2, 24590-WTP-RPT-MGT-14-023, Rev. 2, 2017.

We put science to work.™