Developing Waste-Driven Design Requirements for Filtration and Ion-Exchange at the Hanford LAWPS Facility – 16394

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ABSTRACT

The United States Department of Energy is building the Low Activity Waste Pretreatment System (LAWPS) at the Hanford site near Richland, Washington to treat liquid tank waste. The primary unit operations are filtration for solids removal and ion-exchange for cesium-137 removal. The properties of the stored waste are diverse, therefore the design requirements need to allow for a variety of feeds from the Hanford tank farms. However, it is not cost effective to allow a few rare feeds to define the requirements when there are other options for treating the rare wastes. The waste-derived design requirements were based on trends from this data. The primary feed requirements that will drive filtration and ion-exchange performance are solids content of the feed, cesium to sodium ratio, and potassium concentration. Additionally, aluminum and total organic carbon (TOC) concentrations are considered for flammable gas generation. For the cesium and potassium concentrations, the available experimental data was collected, sorted, and compared against the historic requirements. The design basis for cesium and potassium feed parameters developed here enveloped all but a couple outlier tanks. A review of available grab sample data indicates that occasionally the samples would have up to 0.8 wt% solids concentration. One single sample had 3.3 wt% solids concentration. These solids concentrations were assigned as the nominal and upper bounding solids concentrations. Flammable gas generation does not directly impact the ion exchange process throughput but does impact the design safety requirements, thus TOC and aluminum concentrations (which are inputs to the flammable gas calculations) were examined in a similar way, but were paired together through a simplified correlation to reach an upper bounding value of 0.6 wt% $TOC*Al^{0.4}$.

INTRODUCTION

For the startup of the Low-Activity Waste (LAW) Vitrification Facility at the Waste Immobilization and Treatment Plant (WTP) with the direct feed of low-activity waste (DFLAW), a new process to separate cesium and solids from the sodium-bearing waste is being designed. The Low-Activity Waste Pretreatment System (LAWPS) is designed to remove cesium and solids from the liquid tank waste and transfer it to WTP LAW Vitrification facility. The DFLAW plan includes starting to process liquid sodium-bearing waste to free up tank space while waiting for the WTP pretreatment

facility (PTF) to treat all waste. Given that LAWPS is not required to treat all waste, the LAWPS facility can be more stringent in what waste it can accept.

The LAWPS facility has two primary waste treatment unit operations: filtration, and ion exchange. The selected technologies are cross-flow filters, and spherical resorcinol formaldehyde (sRF) resin (Murkes and Carlsson, 1988; Hassan et al., 2014; Fiskum et al., 2006a and b; Brown et al., 2011). The beginning of the conceptual design of the LAWPS used the same technologies as the PTF so that experience can be gained by applying them. The resin is selective for alkali ions with the hydronium ion (H^+) being most preferred ion due to the resin's weak acid nature (Gorshkov et al., 1998). Cesium has higher preference than potassium on the resin, and potassium has higher preference over sodium (Nash and Isom, 2010) because of the ability of potassium and cesium to dehydrate on the resin (Gorshkov et al., 1998).   Given that sodium and potassium can compete with cesium for the resin, the concentration of potassium and cesium in the feed are important design parameters with sodium being a set constraint. The purpose of the present study is to report the optimal design-basis waste properties for use in crossflow filtration and ion-exchange design.

The primary waste properties that define the filter sizing are the waste liquid viscosity and the solids content (Murkes & Carlsson, 1988). The likely range of solids concentrations in LAWPS feeds is evaluated here. The waste viscosity is somewhat correlated to the sodium molarity, and early work has shown that sodium molarity can often substitute for viscosity in empirical filtration models (Geeting et al., 2005; Foust and Ghosehajra, 2010). The sodium molarity targets are set at LAWPS by system flowsheets, and the feed can be diluted to any target sodium molarity.

Hanford waste generates flammable gas; thus the flammable gas generation rates are an important input to safety design calculations. Hanford waste generates flammable gas both from radiolysis and thermolysis of organics. The radiolysis rates depend largely on the concentration of cesium-137. Models of thermolysis rates, however, indicate that they depend on both the organic carbon and aluminum concentrations (Hu, 2012).

FEED SOLIDS CONTENT

The feed to the LAWPS facility will be double-decanted prior to being fed to the facility. This double-decantation process would be expected to allow the overwhelming majority of the solids in the feed to settle. The early design specification for the LAWPS assigned a solids concentration of the feed to 10 weight percent (wt%), a value that came from a design guide for slurries in the Hanford tank farm. Given that the overwhelming majority of the solids would have settled

from the feed prior to delivery, 10 wt% solids is excessive. Thus, new values for the solids content of the feed were developed.

Based on experience with previous waste transfers, a small amount of entrained solids may be transferred with supernatant. An earlier study compiled all of the data available on solids content of grab samples of supernatant layers in Hanford tank waste soon after slurry transfers (Haigh, 2011). The majority of those samples had no visible entrained solids. Occasionally, the lab would describe some samples as having a trace amount of entrained solids. The lab could not quantify the amount of solids when it was less than 2 volume percent (vol%), so those samples with trace amounts of entrained solids were described as having less than 2 vol% solids. Consequently, setting the nominal (for design) solids content at 2 vol% solids would be conservative. The conversion factor used for solids concentration is 2.5 vol% per wt% (Haigh, 2011). This converts 2 vol% to be 0.8 wt%. Thus, 0.8 wt% solids was used as the nominal solids content for cross flow filter sizing.

As a rare occurrence, one sample from tank 241-AW-102 was reported to be 8.3 vol% (3.3 wt%) solids in a surface sample (Hall 2007; Haigh, 2011). This larger solids content only occurred in one sample out of many and was thought to be rust particles rather than from the waste. For the majority of samples taken for supernatant, no visible solids or less than 2 vol% solids was reported. Consequently, 3.3 wt% is a conservative upper-bounding solids concentration.


ION EXCHANGE

The original design basis cesium-137 concentration used for conceptual design was 0.5 Ci/L. The limit was originally taken from other facilities, so this study investigated if this was a reasonable cesium-137 concentration to use for LAWPS preliminary design basis.   There is a wide range of resin bed sizes that can be adequate for treating a wide range of feed compositions. There are trade-offs in resin bed sizing that must be considered, and the sodium, potassium, and cesium ratios only play a partial role in this consideration. The throughput through the resin is expected to be relatively insensitive to the size of the resin bed when waste is being treated. Resin bed size determines the frequency of resin replacement, capital costs, and frequency of elution and regeneration cycles. A larger resin bed allows for less frequent elution and regeneration cycles so will result in higher throughput over the entire cycle because more time is used treating waste and less time is used eluting and regenerating the resin. However, larger resin beds have major disadvantages because they require larger capital costs for the resin columns themselves, as well as the reagent tank and cesium product tanks. The present study determined if the specification value represented the predominant amount of the waste.

Data was taken from the Hanford Tank Waste Information Network System Best Basis Inventory (BBI) for concentrations of potassium, sodium, and cesium for East Area Double Shell Tanks (DSTs). The reason that East Area DSTs are used is because these tanks are closest to the LAWPS, and thus, most likely going to represent the early feed. The fidelity of the data in the BBI is variable, so the accuracy of the estimates in some tanks is better than others. The purpose of this study is not to definitively obtain the bounding value for the tank waste. The purpose of this study is to determine the upper and lower bounding ratios for the preponderance of data, ensures that these specification values are not be affected appreciably by an inaccurate value for any specific tank by using data from many tanks. Thus, using a wide variety of data serves this purpose, regardless if the data perfectly represents current conditions in any specific tank.

The LAWPS facility is being designed to treat waste over a sodium molarity range between 4 and 6. Most of the waste is currently stored at sodium molarities much higher than this in order to save tank space. However, most waste will be diluted down prior to treatment. When the waste is diluted down the cesium-137 concentration will also be diluted down, so we cannot simply use the current cesium-137 concentrations to evaluate the specification value.

The cesium-137 concentrations in units of Ci/L were calculated by taking the cesium-137 to sodium ratio and multiplying it by the maximum sodium concentration, 6 M. This results in the normalized cesium-137 concentration for the upper end of concentrations expected to be feed to the LAWPS facility, shown in Figure 1. This figure shows that there are two tanks that exceed the specification value and one tank right at the specification value. All of the rest of the tanks are below the specification cesium-137 concentration when normalized to 6 M Na. It should be noted that one tank is more than triple the value in the specification. It does not seem reasonable to design the LAWPS facility to this one single tank when the WTP pretreatment facility will eventually come on line and will be able to treat all waste. Consequently, the specification was kept at 0.50 Ci/L. If the two tanks that are above the specification ever do need to be treated at the LAWPS, the waste planners can either dilute the waste below 6 M Na to get a lower cesium concentration or they can blend the waste with another tank. Thus, there is still a way to treat this waste in LAWPS if ever required.

Figure 1. Cesium-137 Concentration Ranked from Lowest to Highest for Best-Basis Inventory Data Normalized to 6 M Na.
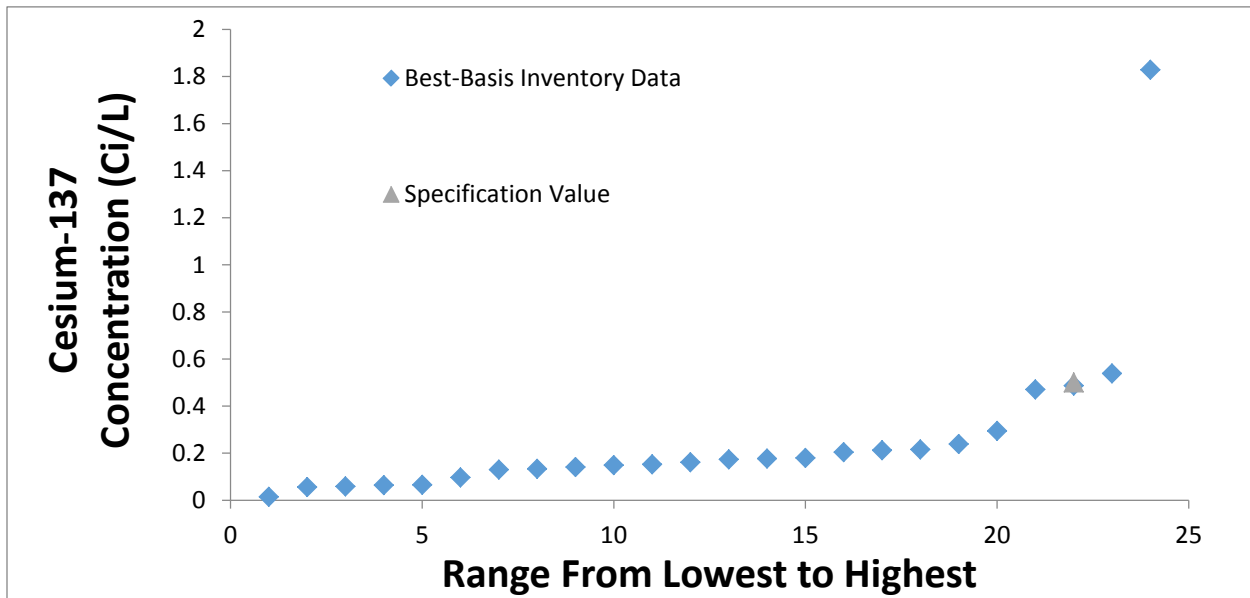
The potassium concentration in the original specification (0.122 M K$^+$) was based on the average potassium concentration in an early feed estimate. Plotting the K$^+$ concentration for all East Area DSTs (interstitial liquid and supernatant) as well as the value for the specification, arranged from lowest to highest confirms that 0.122 M K$^+$ is too low to bound most of the waste (Figure 2). Figure 2 has the original value in the specification in addition to a new proposed bounding value of 0.35 M. Choosing a value of 0.35 M K$^+$ would bound the overwhelming majority of the current tank waste, and some of the tanks with extremely high potassium concentrations may drop below this value if blended with low potassium concentration feed. It should be noted that outlier wastes can still be treated at LAWPS with these recommended bounds, but the resin would have to be eluted more frequently.

The upper bounding values of cesium were needed for sizing of equipment. In the case of potassium, however, the minimum concentration is also important for some safety calculations where one would maximize the available cesium on the ion exchange resin. Therefore, it is recommended that a minimum potassium concentration of zero be used.

Figure 2. Potassium Molarity of East Area Double-Shell Tanks Ranked from Lowest to Highest.
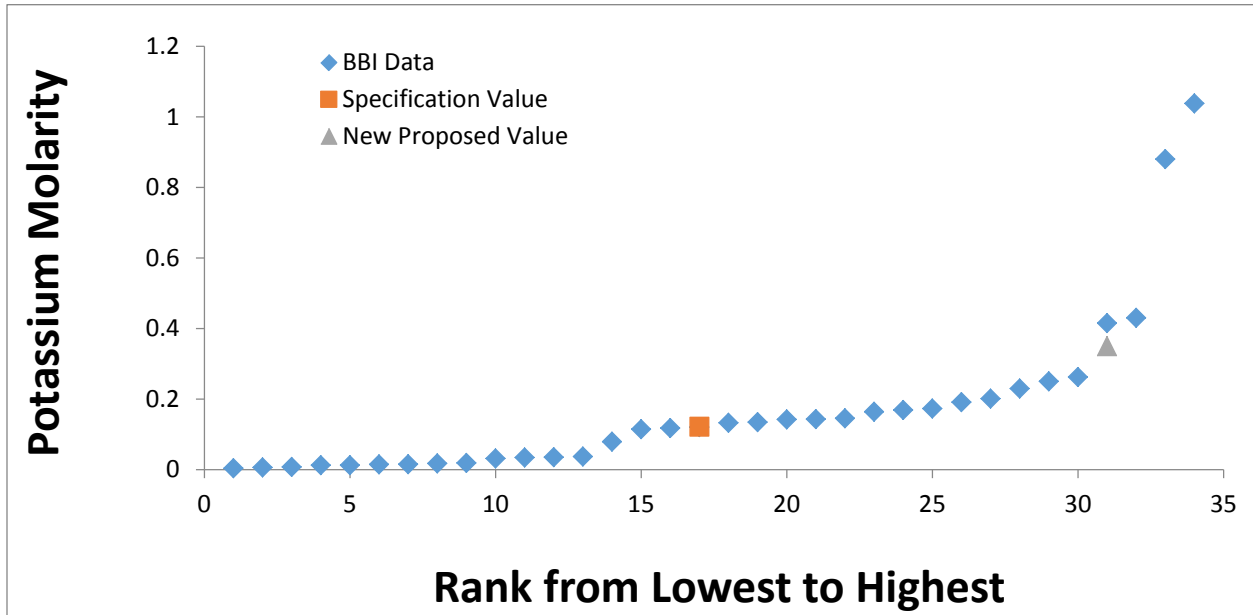

FLAMMABLE GAS GENERATION RATES

While aluminum and total organic carbon concentrations (TOC) do not directly impact the performance of the ion exchange process, models indicate that they do impact the hydrogen gas generation rate in tank waste (Hu et al., 2012). A gas generation calculation is used in safety analysis for the design of the column. The higher the aluminum and TOC concentrations, the higher the thermolytic gas generation rates. The upper bounding aluminum and TOC concentrations are therefore inputs into those calculations. Here, the aluminum and TOC concentrations used to calculate those rates are evaluated. Inspecting the Best-Basis Inventory for east area DST liquid layers indicate that the highest aluminum concentration (normalized to 6 M sodium) is 0.86 M in tank 241-AN-105 and the highest TOC concentration is 0.35 M in tank 241-AW-103. However, it is unrealistic for TOC and aluminum concentrations to be examined separately because the tank with the highest TOC is not the tank with the highest aluminum. Therefore comparing the product of the aluminum and TOC concentration would result in a more realistic limit. Hu (2012) indicates that the thermolysis contribution to the flammable gas generation rate is proportional to the Wt% TOC concentration multiplied by the 0.4 power of the aluminum Wt%. Hence, we defined the limit for aluminum and TOC in terms of $[TOC] \times [Al]^{0.4}$ rather than the individual

concentration of TOC or aluminum. Thus, the $[TOC] \times [Al]^{0.4}$ was calculated for each east area DST and plotted from lowest to highest value (Fig. 3). The highest value came to be 0.58 for tank 241-AW-104 (Figure 3). Due to the increasing trend with no significant outliers (Figure 3), the maximum value was rounded to 0.6 and retained as a design basis.

Figure 3. Product of Al to the 0.4 Power and TOC Concentrations Converted into Wt% and Sorted by Tank Ranking
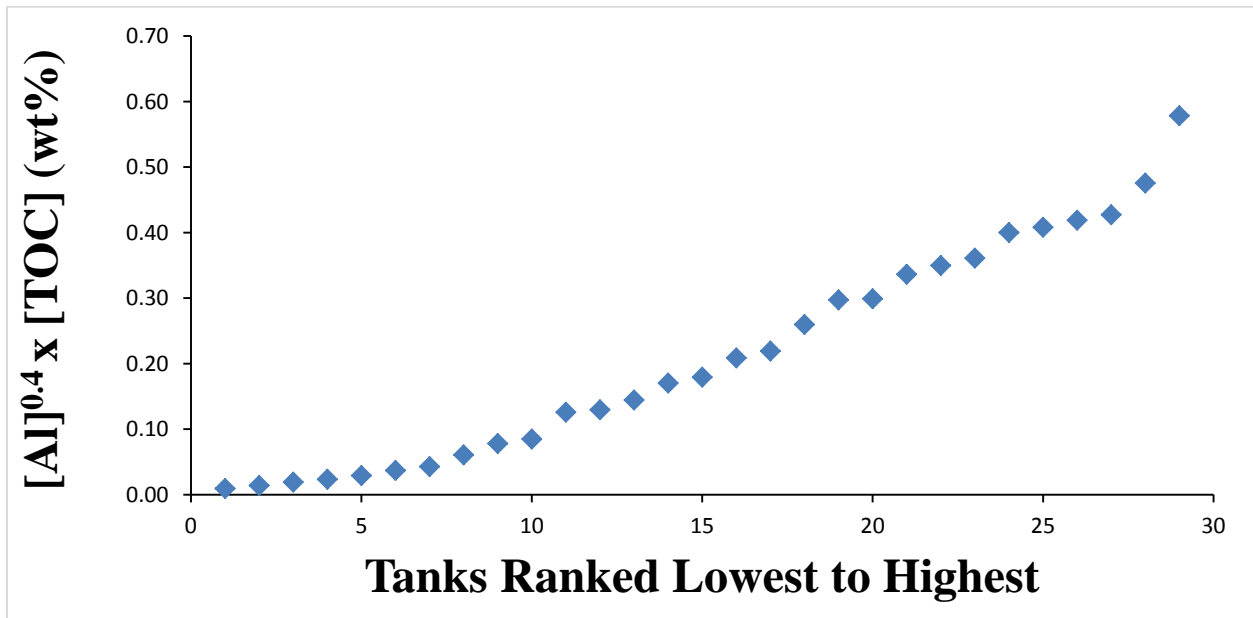
CONCLUSION

This study reports the waste derived design requirements for the LAWPS facility. With these requirements, the most extreme cases of waste are excluded from the design of LAWPS allowing the facility to treat the majority of supernatant in tank farms without being excessive. This study recommends the lower bound of zero moles/L be used for potassium. The rest of the recommendations are upper bounds, and shown in Table 1.

Table 1: Summary of Upper Bound Requirements

Species	Upper Bound
Solids	3.3%
Aluminum	0.6[1]
TOC	
Cesium-137	0.5 Ci/L

Potassium	0.35M
Sodium	6M

[1]Value is without units due to being the product of values of wt% (mass fractions)

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