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Maximizing Production Capacity from an Ultrafiltration Process at the Hanford Department of Energy Waste Treatment Facility

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Abstract: The Department of Energy has contracted Bechtel National, Inc. to design, construct, and commission a Waste Treatment and Immobilization Plant (WTP) to treat radioactive slurry currently stored in underground waste storage tanks. A critical element of the waste treatment capacity for the WTP is the proper operation of an ultrafiltration process (UFP). The UFP separates supernate solution from radioactive solids. The solution and solid phases are separately immobilized. An oversight review of the UFP design and operation has identified several methods to improve the capacity of the ultrafiltration process, which will also improve the capacity of the WTP. Areas explored were the basis of design, an analysis of the WTP capacity, process chemistry within the UFP, and UFP process control. This article discusses some of the findings of this oversight review in terms of sodium and solid production, which supports the treatment of low activity waste (LAW) associated with the facility, and solid production, which supports the treatment of high level waste (HLW) associated with the facility.

Keywords: Mixed waste, process optimization, optimal endpoint solids concentration, ultrafiltration, optimal design

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INTRODUCTION

The Department of Energy's (DOE) Hanford site currently stores the largest quantity of radioactive high level waste of any DOE facility (55 million gallons in 177 underground storage tanks). The Bechtel National, Inc. (BNI) is constructing a Waste Treatment Plant (WTP) to treat and immobilize this tank waste. The WTP is a treatment plant that turns radioactive slurry into glass and involves several distinct stages to fulfill this task. The first area of the plant is pretreatment that dewateres the received radioactive slurry and separates the waste into a high level and low activity waste. The low level waste, or low activity waste (LAW), is further treated to remove radio-cesium before conversion to glass. The high level waste (HLW) is washed and concentrated and converted to glass in a HLW Vitrification facility. A critical element of the pretreatment processing is the UFP (see Fig. 1). The treatment capacity of the WTP is measured in terms of sodium production, which is a direct measure of the amount of LAW glass produced and solids production, which is a direct measure of the amount of HLW glass produced.

The purpose of this research is to determine the conditions that will support the maximization of sodium and solid production. Ultrafiltration is a membrane separation process that uses physical exclusion of material by a porous membrane of a certain pore size and a pressure gradient. The intent is that particles below this pore size and liquids freely pass through the pores. This is termed removal efficiency. As the ultrafiltration process continues, a cake layer may form which reduces the effectiveness of the ultrafiltration process in terms of permeate fluxes but may in fact improve removal efficiencies. This method is used at Hanford to separate LAW, which passes through the membrane, and HLW, which is retained by the membrane and recirculated to the slurry holding vessel.

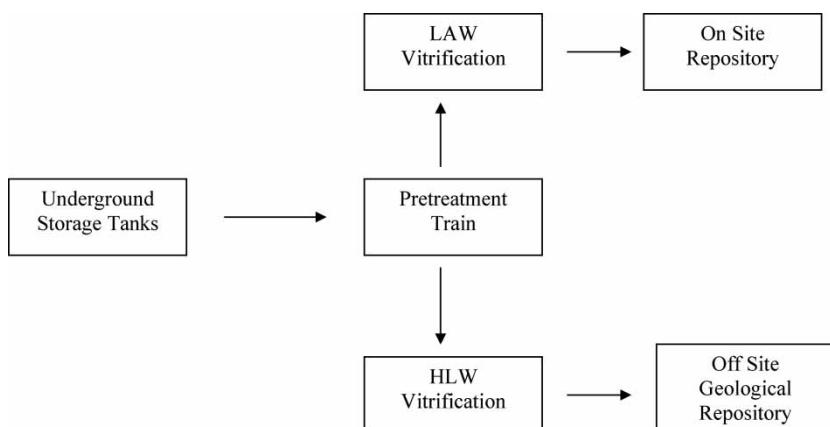


Figure 1. Process flow diagram for waste treatment plant.

The current method of separation is to run a process for as long as it is practical to do such, which for the radioactive slurries generally encountered at Hanford is 20% weight solids (Cs). Another approach advocated in this research is to run the process to an optimal endpoint solids (Cs*) concentration that will maximize the capacity of sodium and solid production (10).

The Hanford inventory of radioactive waste slurry has been delineated into four envelopes with distinct chemical and rheological properties (1, 2). These envelopes are A, B, C, and D, where A, B, and C are liquid phase and D is a solid phase material. Most of the inventory at Hanford is envelope A. Only a couple of the 177 underground storage tanks have been designated as envelope C material.

Given in Fig. 2 is a process flow diagram for the UFP, which includes 3 membrane bundles in two trains (one train depicted). There are two banks of UFP trains being proposed with a membrane surface area of 710 ft², recirculation flow rate of 2200 gpm, and transmembrane pressure differential of 50 psi.

The outline of this paper is to discuss previous work on the determination of an optimal endpoint solids concentration for ultrafiltration; discuss slurry properties associated with treatment using the UFP; present a sodium analysis; present a solids analysis; for solid production, show that a relationship exists between membrane surface areas and Cs*; where Cs* is the optimal bulk phase solids concentration to maximize solids production; and discussion and conclusions.

BACKGROUND

A small body of literature exists to support the idea that ultrafiltration has an optimal endpoint concentration of solids to minimize the process time (maximize capacity). This literature includes (5, 6, 9). The existing literature assumes a form of the film model is applicable (4) to describe the flux rate over time and uses this model as the starting point to determine an optimal endpoint solids concentration; the development of the film model assumes a constant

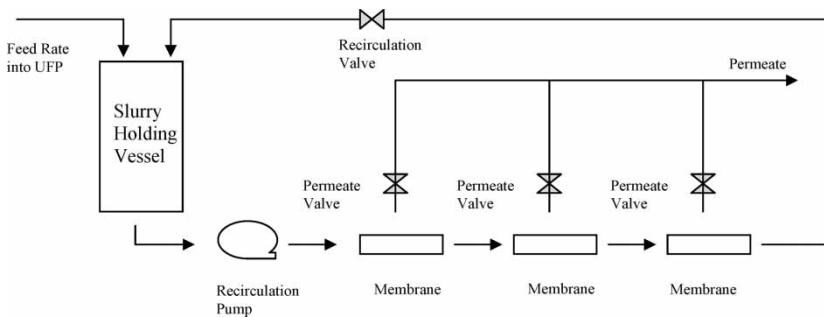


Figure 2. Process flow diagram for ultrafiltration process.

concentration at the wall and a mass transfer coefficient that does not change with time.

For the UFP system studied at Hanford, this model would be in error due to the long time required until a steady state cake layer develops and that the fluid properties of the slurry change over the course of the operation. (5) and (6) have incorporated osmotic pressure and viscosity dependence on the mass transfer coefficient, but assumed negligible membrane fouling, which is significant for the UFP. The chemistry, fluid properties, and rheology associated with the UFP are likely to preclude a satisfactory analytical approach to finding an optimum for the endpoint solids concentration and as such, an empirical approach has been utilized in this paper.

The typical approach to determine the optimal endpoint solids concentration in the literature is to find the solution to the following optimization problem

$$\frac{dt}{dC_b} = 0 \quad (1)$$

where t is the time to dewater and C_b is the concentration of solids in the bulk phase. In this paper, an optimum endpoint solids concentration is presented to maximize the capacity of annual sodium production and daily solids production at various membrane surface areas, slurry property scenarios, and busy times.

Slurry Properties

Given that only a few of the 177 UST within the Hanford inventory are envelope C this analysis will ignore these few tanks and consider the inventory as 90% envelope A/D and 10% envelope B/D; Envelope B/D is represented by Geeting's curve and is given from (8). The characterization of envelope A/D permeate flux is given from (3). Model results for permeate flux curves from the above experiments are presented in Fig. 3.

The solid analysis was conducted under the assumption that the entire Hanford inventory was envelope B/D. The correctness of this assumption has been investigated within the sodium analysis by conducting the analysis under two scenarios: assume the entire inventory is envelope B/D (Geeting Scenario) and assume the inventory is 90% envelope A/D and 10% envelope B/D (Hanford Scenario), which is more representative of the Hanford inventory.

Consideration of slurry properties is provided to determine its effect on maximizing capacity. It will be shown that there is no apparent effect on when to stop the process (C_s^*), but the mass of sodium produced is obviously higher for a material with a higher flux rate. Thus, representing the slurry as envelope B/D (Geeting scenario) versus 90% envelope A/D and 10% envelope A/D (Hanford scenario) reduces the annual sodium production.

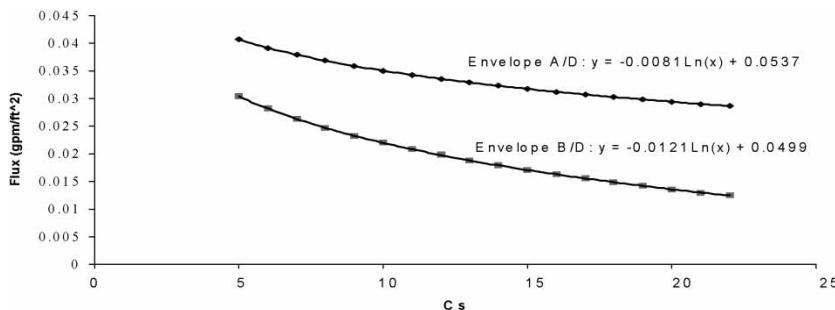


Figure 3. A/D and B/D permeate flux curves.

Sodium Analysis

Most of the production of sodium occurs during the dewatering phase of ultrafiltration. Sodium freely passes through the membrane and it is likely that maximizing the capacity of the solids production will also maximize the capacity of the sodium production.

Several stages occur within the operation of the UFP where the duration and necessity of each stage is dependent on the material being treated. The first stage is a dewatering stage where the solids are retained by the membrane and microsolute and liquids freely pass through the membrane and are conveyed to the cesium ion exchange toward LAW vitrification. The next step is the removal of sodium and other microsolute by a solids washing, which is accomplished by adding a sequence of 1000 gallons of process condensate while the UFP is running to extract these microsolute; this is followed by a caustic leach. The caustic leach maintains aluminum and other metals in suspension, which is followed by a second solids wash. The final stages include sampling of the material, transfer solids, and ultrafilter cleaning. The various stages of operation for the UFP are given in Fig. 4.

Reducing the time to complete an ultrafiltration run is best accomplished by reducing the dewatering time because this stage of the UFP processes represents the bulk of the process time. Reducing process time for a given mass of solids improves capacity.

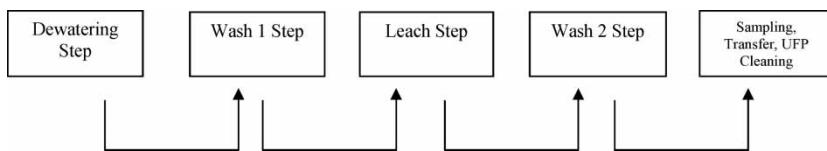


Figure 4. Stages of Hanford ultrafiltration.

The capacity of the UFP system was evaluated for a range of final solids concentrations that varied from 5 wt% to 25 wt%. In all cases a feed concentration of 3.75 wt% and an initial slurry volume of 22,000 gallons was used.

The first step was to determine an average permeate rate over the range of insoluble solids considered, which is mathematically expressed as

$$Q_p = \frac{1}{2}(\gamma_{3.75} + \gamma_{Cs})A \quad (2)$$

where Q_p is the average permeate flow rate (gpm), $\gamma_{3.75}$ is the permeate rate associated with 3.75 %weight solids, γ_{Cs} is the permeate rate associated with the endpoint %weight solids, and A is the membrane surface area. Three surface areas were considered: 592 ft², 710 ft² and 886 ft².

To determine the volume of permeate,

$$V_p = Q_p(t_{Cs} - t_{3.75}) \quad (3)$$

where V_p is the permeate volume (gallons), Q_p has previously been defined, t_{Cs} and $t_{3.75}$ are the times at the endpoint Cs and %weight solids of 3.75 given from empirical relationships in Fig. 5 for Envelope A/D. To determine the metric tons of sodium produced per a batch,

$$Mass = 5 \frac{moles}{l} * V_p \quad (4)$$

The result is given in Fig. 6 for envelope A/D. An identical analysis was conducted for envelope B/D.

The total time to run both trains of the UFP was determined, given that the trains do not run fully in parallel, is either twice the time of dewatering if this time is longer than the time for the other stages or the total time is twice the dewatering times and the sum of the times for the other stages. The cut-off condition is determined by the following equation:

$$2 * T_{Conc} \geq T_{Wash} + T_{Leach} + T_{Transfer} + T_{Clean} \quad (5)$$

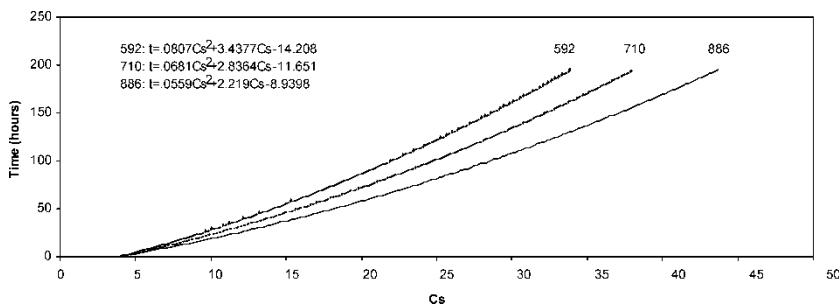


Figure 5. Cs vs. dewatering times.

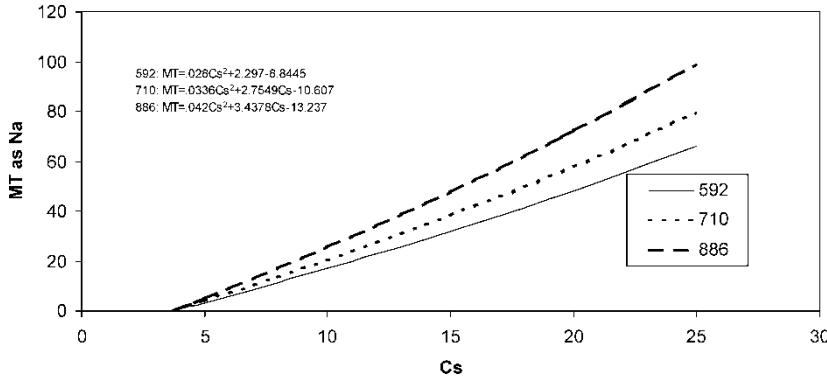


Figure 6. Cs vs. MT as Na for various filter surface areas.

where T_{conc} is the time to dewater, T_{wash} is the wash time, T_{leach} is the leach time, $T_{transfer}$ is the time to transfer slurry, and T_{clean} is the acid wash time (T_{leach} , $T_{transfer}$, and T_{clean} are assumed constant).

If the condition holds,

$$T(cycle) = 2 * T_{Conc} \quad (6)$$

else

$$T(cycle) = 2T_{Conc} + T_{Wash} + T_{Leach} + T_{Transfer} + T_{Clean} \quad (7)$$

The methodology determined empirical relationships between cycle times to complete two batches for a given endpoint solids concentration (which is defined as Equation (5), and determine the number of batches that could be completed for an operation year, which is defined as

$$\text{Batches/Year} = \text{Utility Time} * 24 \text{hours/day} * 365.4 \text{ days/Cycle Times} \quad (8)$$

Such that annual production is then

$$\text{Annual Sodium Production} = \text{Batches/Year} * \text{MT/Double-Batch} \quad (9)$$

Figures 7 and 8 give the results of this analysis for 90% and 70% attainment time for two sets of slurry properties. The first scenario assumes the entire inventory is envelope B/D (Geeting Scenario) and the second scenario assume 90% of the inventory is envelope A/D and 10% is envelope B/D (Hanford scenario).

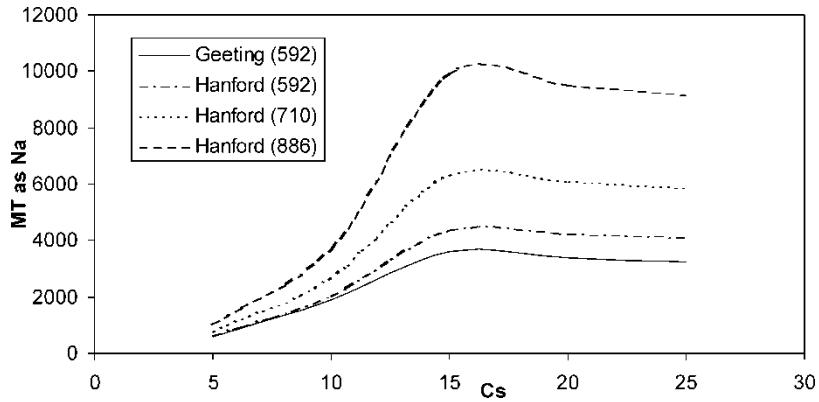


Figure 7. Annual sodium production at 90% attainment time.

Solids Analysis

A mass balance equation was developed for the metric tons of solids produced per a day

$$W_s = \frac{V_{UFP} C_s \rho_s \rho_f S_L}{C_s \rho_f + (1 - C_s) \rho_s} \frac{1}{t} \quad (10)$$

where W_s is the solids transfer rate in MT/day, V_{UFP} is the volume of the UFP feed vessels, C_s is the final solids concentration in the UFP vessel, ρ_s is the density of solids fraction of slurry, ρ_f is the density of the fluid fraction of the slurry, S_L is the fraction of solids remaining after washing and leaching, and t is the cycle time to complete a full cycle on both trains. The leach/wash factor (S_L) varied from .4 to .7. During the wash and leach stages it is likely to lose solids and this is accounted for by the use of leach/wash factors.

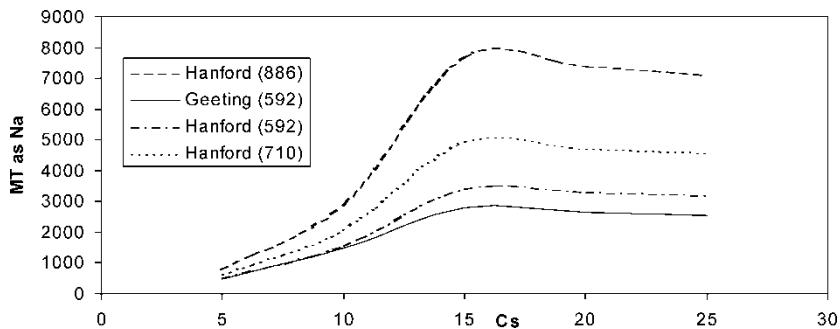


Figure 8. Annual sodium production at 70% attainment time.

Other mass balance equations account for the concentration of solids in the UFP feed vessel as the slurry concentrates and were developed from relationships of permeate flux rate per time given in Fig. 3 assuming all material is envelope B/D (6). This is a conservative assumption and under estimates the actual production levels (see complementary article). Empirical relationships for dewatering times and wash times per a membrane surface area were then derived from these mass balance equations and are given in Figs. 5 and 9. These relationships were developed by keeping the feed vessel at a constant volume (22,000 gallons) by adding a feed stream of 3.75% weight solids from a feed prep vessel at the same rate as the permeate is drawn from the UFP.

The time required to complete the dewatering stage of the process is most heavily affected by the assumption on final concentration as shown in Fig. 5. The times for the other stages are practically independent of the final concentration (10). For the lower concentration limits (e.g., <10 wt% to 15 wt%) the dewatering time is shorter than the time required to complete the other stages. Accordingly, the dewatering cycle in the second loop is completed before the washing, leaching, and cleaning stages are complete in the first loop. As soon as those stages are complete, therefore, the first loop will start its dewatering cycle. For the higher concentrations this is not the case. The first loop finishes all stages of the cycle before the second loop has completed its dewatering stage. The first loop, therefore, has a wait time before starting its next dewatering cycle. These characteristics of the operation of the loops were considered in evaluating the capacity of the system. The time was determined using the method given above and expressed as Equations (5), (6), and (7).

Given Equations (8), (9) and density of the solid and liquid fractions of the slurry, the results of the solid production for membrane surface areas of 592 ft², 710 ft², and 886 ft² are given in Fig. 10, which is discussed below.

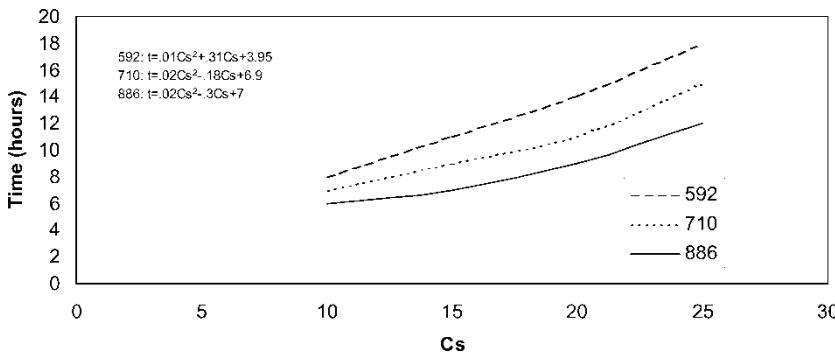


Figure 9. Cs vs. wash times.

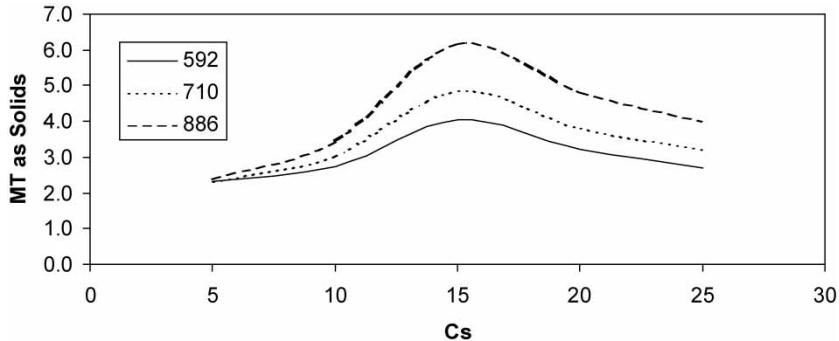


Figure 10. Daily solids production.

Figure 10 shows the calculated solid production capacity of the UFP system in MTG/day equivalent as a function of the final concentration of % weight solids. This figure shows that the system peak capacity in terms of solid production is a maximum for a solids concentration of 15% weight solids. This maximum occurs because of

- (1) the non-linear relationship in permeate flux rate with increasing bulk concentration during the dewatering stage for higher concentrations, and
- (2) the longer time required to complete the water washing, leaching and cleaning stages of the cycle compared with the shorter dewatering time for the lower concentrations.

As one would expect, a larger surface area produces more solids per a unit time. It will be shown in the next section that surface area effects CS^* with a larger membrane surface area having a larger CS^* .

Investigation of the Relationship Between CS^* and Membrane Surface Area

As stated in the introduction and shown in Fig. 10, an optimal endpoint Cs exists to minimize the operation time (maximize capacity) in terms of solids and sodium. As much of the sodium is produced during the dewatering phase, this optimum should be the same for both solids and sodium. It is shown in Appendix A that an optima occurs when

$$\frac{\partial W_s}{\partial Cs} = 0 \quad (11)$$

where W_s is the daily production of solids Equation (10) and Cs is the endpoint solids concentration. The optima is expressed by the

following equation:

$$Cs^* \approx \sqrt{\frac{c + T}{.85a - .45b}} \quad (12)$$

Using the empirical relations developed in Fig. 11, $T = 58.5$ hours, and Equation (12), the following optima were determined

As shown in Table 1, Cs^* is dependent on membrane surface area with an increase in Cs^* at increased membrane surface areas where the purpose of this section was to demonstrate that a relationship exist between membrane surface area and Cs^* . The values developed in Table 1 are from a stochastic approach and are only estimates of the actual Cs^* at a particular membrane surface area.

In (7), it was proven that the optimal mode of operation in terms of feed rate into the UFP and transmembrane pressure differentials is to match the rate of material leaving as permeate with material coming into the UFP (permeate matching diafiltration). An empirical model for the time to dewater, which is time optimal in regards to dewatering times, is

$$T^* = \frac{-1}{k} \ln \left[1 - \frac{k}{a} (C_f - C_0) \right] \quad (13)$$

where T^* is the optimal time, k is a parameter of the material being treated, a is a constant, C_f is the final bulk phase concentration, and C_0 is the initial bulk phase concentration.

Further, the above equation can be manipulated into the following form

$$\frac{q_0}{k} \frac{1 - e^{-kT^*}}{(C_f/C_0 - 1)} = \frac{V}{A} \quad (14)$$

where $\{q_0, k\}$ are parameters of the material being treated, V is the volume of material to be treated, and A is the membrane surface area.

And it is easy to show that increasing the membrane surface area increases C_f , which is also Cs^* .

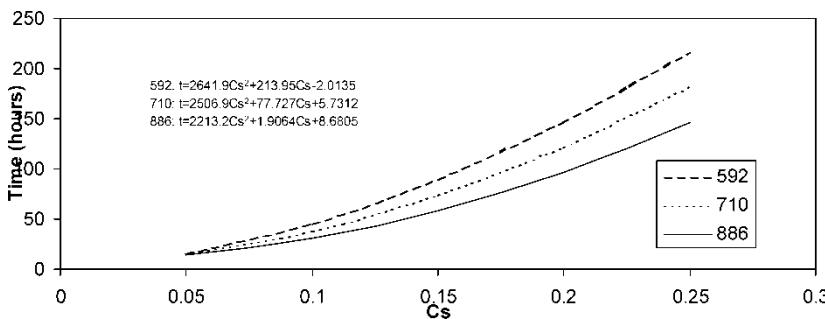


Figure 11. Cs vs. time for dewater + wash.

Table 1. Optima for solids operations

Filter surface area (ft ²)	Optima (% weight solids)
592	16.2
710	17.5
886	18.9

DISCUSSION

Sodium Analysis

As can be seen in Figs. 7 and 8, an optimal endpoint concentration of solids is seen for each case considered. It is observed that the optimum is around 15% weight solids. Also, including permeate rates for envelope A/D has improved production by 20%.

The reason that an optimum endpoint solids concentration to maximize annual sodium production exists is that there is a trade-off between the time to complete a batch of a given mass and the amount of batches that can be processed in a year. Ending the process at a lower C_s allows more batches per a year but at a lower mass per batch; ending the process at a higher C_s allows fewer batches per a year but at a higher mass per batch.

An optima was shown in maximizing solids production as a result of the nonlinear relationship of the cycle time and the developed mass balance equation for daily solids production, which is a similar form of optima to that found in the sodium analysis.

As previously mentioned, the sodium analysis was conducted under two scenarios: assuming the entire Hanford inventory is envelope B/D and a scenario where 90% of the inventory is envelope A/D and 10% is envelope B/D, which is more representative. Under each scenario, the optimum for a given membrane surface area does not appear to be dependent on the slurry properties.

Another interesting feature to Figs. 7 and 8 is that the production of sodium at a concentration of weight solids at or above C_s^* has been observed to be proportional to the square of the membrane surface area, e.g.,

$$MT \propto kA_m^2 \quad (15)$$

where MT is the annual metric tons of sodium produced, A_m is the membrane surface area, and k is a proportionality constant. A linear relationship was observed for solids production when the concentration of weight solids was at or above C_s^* , which is 15% weight solids. No apparent reason for these observations has been determined and this will be the subject of future research.

Solids Analysis

All membrane surface areas produce the same amount of solids to a given endpoint solids concentration; the benefit of more surface area is in how fast the process can dewater to a final % weight solids. An optimum appears to exist because the total cycle time for a double batch of slurry to a final % weight solids is not a monotonically increasing function but nonlinear due to the inequality defining T_{cycle} given in Equation (9) and that the mass of solids produced Equation (8) is nonlinear. The nature of this optimum is similar to the optima found for the maximizing sodium production capacity where a trade-off condition exists between the mass of sodium produced versus the time to produce the sodium; where ending the process at a higher endpoint solids will produce more sodium but at a longer process time versus ending the process at a lower endpoint solids, which produces less sodium but at faster processing times.

It has further been determined that the optima is dependent on membrane surface area; an increase in membrane surface area results in an increase Cs^* . The reason for this relationship is the subject of future research and no apparent reason has been determined at this time. Future research will validate this observation by showing that the optimal endpoint of Cs for the three membrane surface areas considered in this paper are slightly different.

Ongoing research to improve the performance of the UFP at Hanford includes the development of a dynamic model of the UFP amendable to control theory, assessing the benefits of an advance process control method known as optimal control, and comparing simulation results in this paper and ongoing research against empirical results of a bench scale apparatus. Another interesting research area is to investigate the relationships between Cs^* , membrane surface area, and production levels; it needs to be emphasized that several methods can be combined to improve the capacity for solids (and sodium) production to include form of process control utilized, endpoint Cs , and surface membrane area utilized. These parameters of the system interact and the interaction must be addressed when assessing and improving performance of the UFP.

CONCLUSIONS

It has been demonstrated that an optimal endpoint solids concentration to maximize sodium production exists; this analysis was conducted based on the mass balance equations for the system and is more of an empirical approach than has been previously done in the literature.

To maximize sodium production at a given surface area, there is a specific endpoint solids concentration, which has been shown to be 15% weight solids. Another conclusion is that Cs^* is likely not dependent on the particular envelope treated. It needs to be noted that the permeate rate over a given

range of Cs was determined from the two endpoints and a better model may be to include the mid-range as well. This consideration will be addressed in future research.

To maximize solid production at a given surface area, there is a specific endpoint solids concentration in the vicinity of 15% weight solids with consideration for the dependence between Cs^* and membrane surface area where a larger membrane surface area results in a higher Cs^* . To maximize the production capacity of both solids and sodium, an endpoint of 15% weight solids should be utilized, which is dependent on the membrane surface area and may change at higher (or lower) membrane surface areas not considered in this research.

As mentioned above, the assessment and improvement of an ultrafiltration process involves several parameters that interact. This interaction is the focus of ongoing research and must be addressed to assure an improvement in fact results.

APPENDIX A: A VS. CS RELATIONSHIP

The purpose of this appendix is to determine an optima for Cs^* , which demonstrates a relationship between Cs^* and membrane surface area.

$$\frac{Ws}{V_{UFP}} = \frac{U}{V} \omega \text{ and } \frac{\partial Ws}{\partial CS} = 0 = \left(\frac{U}{V}\right)' \omega + \left(\frac{U}{V}\right) \omega \quad (16)$$

$$\frac{\partial Ws}{\partial Cs} = 0 \quad (17)$$

where Ws is the daily production of solids Equation (8) and Cs is the endpoint solids concentration. The derivation given below determines the optima based on Equation (8) and an empirical relationship between Cs and t

$$t = aCs^2 + bCs + c + T \quad (18)$$

where t is the cycle time and T is the time for leaching, cleaning, and transfer.

Where

$$U = Cs\rho_s\rho_f, V = Cs\rho_f + (1 - Cs)\rho_s, \text{ and } \omega = \frac{1}{t} \quad (19)$$

$$\frac{t'}{t} = \frac{U'V - UV'}{UV} = \frac{\rho D}{Cs} \frac{1}{V} \quad (20)$$

$$\frac{t'}{t} = \frac{\rho_s}{Cs} \frac{1}{Cs\rho_f + (1 - Cs)\rho_s} = \frac{K}{Cs[K + (1 - K)Cs]} \quad (21)$$

where

$$K = \frac{\rho_s}{\rho_f} \text{ and } \theta = \frac{1 - K}{K} = \frac{\rho_f - \rho_s}{\rho_s} \quad (22)$$

$$\frac{2aCs + b}{aCs^2 + bCs + c + T} = \frac{1}{Cs + \theta Cs^2} \quad (23)$$

$$2a\theta Cs^3 + (a + b\theta)Cs^2 - c - T = 0 \quad (24)$$

$$- .9aCs^3 + (a - .45b)Cs^2 = c + T \quad (25)$$

$$Cs^2[-.9aCs + a - .45b] = c + T \quad (26)$$

$$.85a - .45b \approx \frac{c + T}{Cs^2} \quad (27)$$

and

$$Cs* \approx \sqrt{\frac{c + T}{.85a - .45b}} \quad (28)$$

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