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Sizing an Ultrafiltration Process that Will Treat Radioactive Waste

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A Department of Energy oversight review has shown that the performance of a waste treatment and immobilization plant at the Hanford Department of Energy facility depends on the performance of the ultrafiltration process utilized. In the present study, a theoretical model for permeate rates using a dead-end filtration theory was used into mass balance equations for the bulk phase concentrations of solids and sodium. Design curves were then generated that show the minimum membrane surface area necessary to treat a certain volume of radioactive slurry in a prescribed time, where the prescribed time could be dewatering time, wash time, or cycle time.

Keywords cross-flow filtration; dead-end filtration; maximizing production and permeate rate study; radioactive waste

INTRODUCTION

This paper supports an effort to understand the performance of an ultrafiltration process (UFP), shown in Fig. 1, which is being designed and built at the Hanford Department of Energy facility. The UFP is within the pre-treatment train, as seen in Fig. 2, of a waste treatment and immobilization plant (WTP). The Hanford Department of Energy facility stores 40% of the nuclear waste inventory of the Department of Energy and this represents 55 million gallons of radioactive waste stored in 177 underground storage tanks (UST). Some of these USTs have begun to leak, and the WTP is being designed and built to remediate this pressing environmental concern.

It is noted in Fig. 2 that there are two products of this plant: canisters of vitrified low activity waste (LAW) and canisters of vitrified high level waste (HLW). A measure of LAW production is the mass of sodium processed; a measure of HLW production is the mass of solids processed.

A proven pinch-point of the WTP is the UFP (1) and improving the performance of the UFP will also improve the performance of the WTP. In this paper, design curves are developed that show the minimum membrane surface area necessary to treat a certain volume of radioactive slurry in a prescribed time. This paper supports an effort to maximize the production of sodium and solids produced from the WTP at Hanford.

This study utilizes a model for permeate rates used in dead-end filtration, and incorporates this model into mass balance equations for bulk phase concentration of solids and sodium. The resulting equations are integrated to find the dewatering time, wash time, and cycle times associated with a given volume of slurry to be treated. Furthermore, since the UFP is a cross-flow system, justification for utilizing a dead-end filtration model is also discussed.

This paper discusses previous research work in this area; develops models for permeate rates, mass balance equation for bulk phase concentration of solids, and mass balance equations for bulk phase concentration of sodium; and presents design curves for dewatering, wash and cycle times.

BACKGROUND

It is agreed in the industry and government that understanding the performance of the UFP will lead to improving its performance and also optimize the performance of the proposed WTP. Previous work in this area includes a Bechtel Design study (2,3), DOE oversight review (1), the revision of permeate models utilized in the DOE oversight review (2,3), and work currently being done by the Pacific Northwest National Laboratory (6–9). Additional studies have been done at other national laboratories (10–12) and similar work (13–16) is being conducted in parallel institutes in other countries such as the Institute of Nuclear Chemistry and Technology, which is in Poland. A brief discussion from these previous studies is presented below.

Bechtel Design Study

Bechtel National, Inc. (BNI) has been tasked with the design, building, and commissioning of the WTP. As part of this effort, they have performed a study (4,5) to determine cycle times for different envelopes of material treated by the UFP. An envelope designates a treatable slurry mixture that has certain chemical or biological characteristics. The three envelopes considered are A/D, B/D, and C

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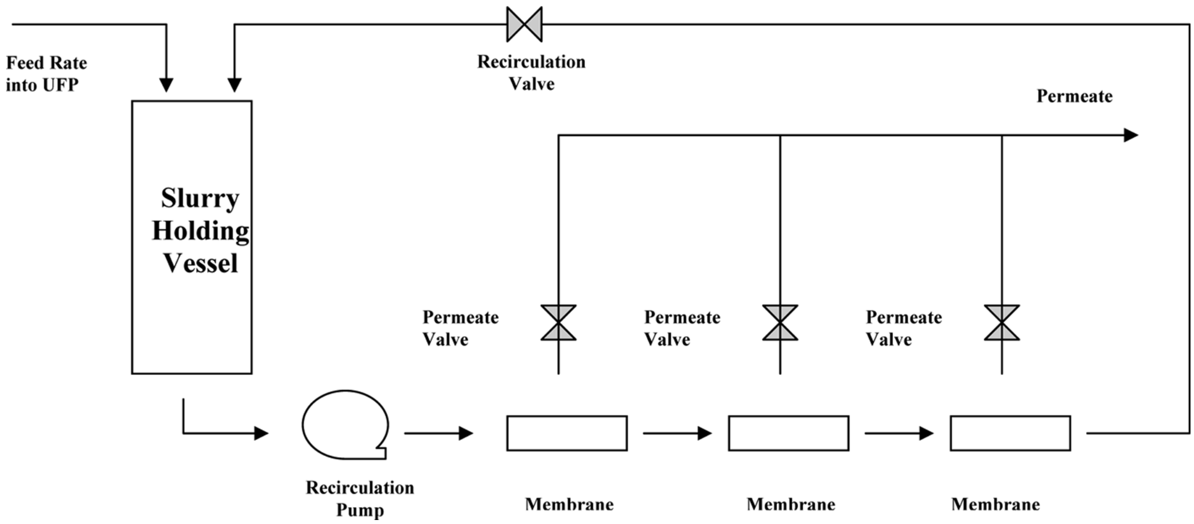

FIG. 1. Ultrafiltration process flow diagram.

where {A, B} designates a liquid component, D designates a solid component, and C is a slurry with organics.

This work is based on the assumption that a regression model is valid for the permeate rates. This regression model is known as the "Geeting's curve" and is based on empirical studies of a stimulant of an Envelope "A/D" material from a 2′ singular, porous tube cross-flow filtration system.

DOE Oversight Review

Several major concerns were found with the BNI design for critical areas of the WTP and one critical area is the performance of the UFP. The DOE Oversight Review (1) conducted a sensitivity analysis of the work done by BNI and tried to remove some erroneous assumptions made. Again, the permeate rate model was Geeting's curve.

Permeate Rate Study

A key assumption of the DOE Oversight Review was that a regression model from a single, 2′ porous tube is valid to a bundle of tubes that 92″ long. This 2′ tube is associated with a bench-scale apparatus known as the Cell Unit Filtration System (CUFS). It is noted that regression models are only good to predict in-sample and do not address issues of physical scale. Another approach (2) was to utilize a model for dead-end filtration and show that it is applicable to the UFP. Foust and Ghosehajra (2) also showed that a regression approach can sometimes give erroneous results, e.g., negative permeate rates.

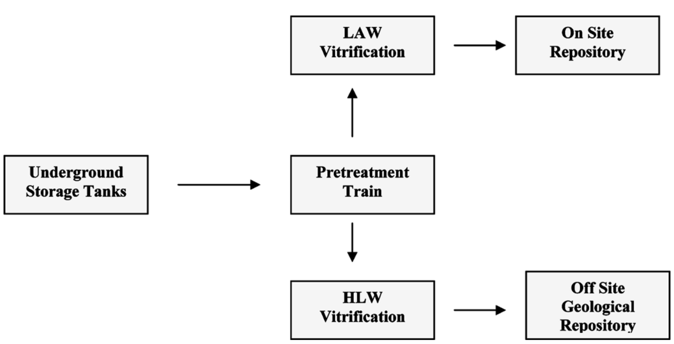

FIG. 2. Waste treatment plant flow diagram.

The permeate rate model in (2) is utilized in the current work.

PNNL Work

The Pacific Northwest National Laboratory has acted as the research arm for both DOE and BNI, and has done many studies on the UFP. Some of the studies applicable to the production of the UFP include (7–9).

In (7), a sample of actual waste was utilized in a pilot scale apparatus and from this study a reliable estimate of permeate rates for actual waste was developed and known as "Geeting's Curve." One concern of this data is that it is for a single, 2′ (.61 m) porous tube and it is unclear how these results will scale-up to a multiple bundled porous tube system where the lengths are 92″ (2.34 m).

In Geeting, Hallen, and Peterson (7), a flux model was developed from two classic models and utilized in Geeting, Hallen, and Peterson (8) where the effects of the pump and control valve settings were gleaned for average filtration rates. This study illustrated that a certain control valve setting for various bulk phase concentration of solids optimizes the average filtration rates.

Recently (9), have looked at a method to characterize the permeate rates for a particular batch of material associated with 177 UST containing radioactive waste (Hanford Inventory). It has been shown that there are "more than

150 different significant sludge-bearing streams" and characterization is a very expensive proposition. This paper delves into utilizing results from centrifuged solid studies to predict long-term (gel concentration) permeate rates.

It is noted that a ¼ scale pre-treatment train is currently being commissioned at Hanford and will help in the understanding of scale issues associated with ultrafiltration applied to radioactive waste, help greatly in how to characterize a particular waste stream, and justify production optimization studies.

SRNL Work

The work of Mark Duignan and his group (11,12) represents a good example of where issues of scale have been more properly addressed. Duignan utilized a 7 full-length tube bundle that addressed issues of length scale that would affect the cake dynamics and also several fluid dynamic issues. This work looked at a stimulant of Envelope C slurry and is utilized as the empirical basis for permeate rates in this paper.

Duignan and Lee (12) have further addressed issues of scale associated with ultrafiltration applied to radioactive waste. This included CFD analysis of the wall shear at the small-scale (CUFS) and pilot scale. The CUFS has often been utilized to provide an empirical basis for the UFP design, but these results are not always discussed in the perspective of scale.

Institute of Nuclear Chemistry and Technology

The work that is being conducted to treat radioactive waste at several U.S. national laboratories is being reflected in other countries at similar facilities. One example is the Institute of Nuclear Chemistry and Technology in Warsaw, Poland. Just as at Hanford different separation processes and polymer additives were explored, so have they been explored in Poland (13–15). More recently, researchers at the Institute of Nuclear Chemistry and Technology are exploring how to optimize the conditions to maximize throughput, which reflects the main aim of the current work (16).

METHOD

The method to develop the design curves presented in this paper involved three distinct stages:

1. Development of permeate rate models
2. Development of mass balance equation for solids
3. Development of mass balance equation for sodium

Once these models were developed and applied to empirical studies for cross-flow filtration, design curves for dewatering, wash, and cycle time were developed.

Permeate Rate Models

This section will develop a model for permeate rates and incorporate that model into Eq. [7]. Once this has been done, a model for dewatering times will be derived and form the basis to Fig. 6, which depicts a minimum membrane surface area for a given set of conditions to include a prescribed dewatering time.

TABLE 1
Pilot scale filtration specifications

Manufacturing and material	Sintered, 316 stainless steel
Length (cm)	229
ID (mm)	12.7
Tubes	7
Nominal Pore Size	0.1 micron
Retention	Roughly 100%
V (m/s)	3.66
TMP (kPa)	276
T (Celsius)	50
Slurry Properties	Simulant of Envelope C

The apparatus utilized is described in Table 1 and it was utilized to treat a simulant to a radioactive waste found at Hanford. The simulant mimics an Envelope C material that includes Tank 241-AN-102 and is composed of organics and other complexants. The simulant is essentially a metal solution at a high pH with a predominant aluminum and sodium chemistry, and lesser amounts of other metals. More details of the slurry and pilot scale apparatus can be found in (10).

In (2,17) it is shown that the filtration processes can be modeled as

$$\frac{t}{V} = aV + b \tag{1}$$

where t is the time to collect a volume of the permeate V and {a, b} are parameters associated with a best-fit line through data from a bench-scale model. The model for the permeate rates is

$$Q_p = \frac{1}{\sqrt{b^2 + 4at}} \tag{2}$$

where

$$a = \frac{\mu \alpha C_S}{2A^2 \Delta p}$$

and

$$b = \frac{\mu R_m}{A \Delta p} \tag{3}$$

where

μ = viscosity of permeate
α = a measure of the compressibility of the cake
C_s = Concentration of solids within the cake
A = membrane surface area

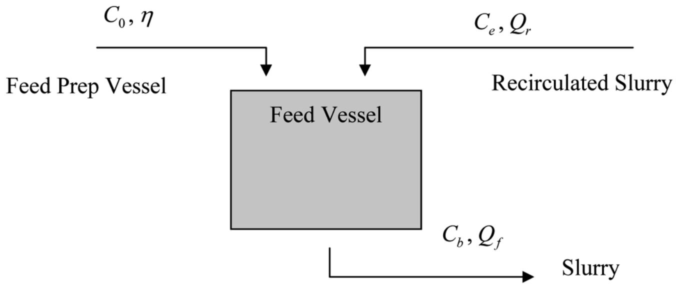

FIG. 3. Mass and flow balances for slurry feed vessel.

ΔP = Transmembrane pressure differential
R_m = Hydraulic resistance associated with the membrane

Several data sets of t/V versus V associated with the UFP at Hanford have shown a high correlation for the linear relationship displayed in Eq. (1) (2).

Mass Balance Equation for Solids

The dynamic model for the UFP relies on the mass and flow balance equations of the slurry holding vessel and membrane tubes. These equations account for a time dependent concentration in the bulk phase (C_b), a time varying permeate model, and a time dependent cake layer, which are presented in Figs. 3 and 4.

The mass balance for this system is

$$\frac{d(VC^S)}{dt} = V\frac{\partial C^S}{\partial t} + C^S\frac{\partial V}{\partial t} = C_0^S\eta \tag{4}$$

and

$$\frac{\partial V}{\partial t} \approx \eta + Q_r - Q_f = \eta - Q_p \tag{5}$$

This results in

$$\begin{aligned} \frac{d(VC^S)}{dt} &= V\frac{dC^S}{dt} + (\eta - Q_p)C^S = C_0^S\eta \\ \frac{\partial V}{\partial t} &= \eta - Q_p \end{aligned} \tag{6}$$

where

C^S = Bulk phase concentration of solids
V = slurry holding vessel volume
C_0^S = Concentration feed of 3.75% weight solids
η = volumetric rate into UFP
Q_p = permeate rate

For constant volume operations, Eq. (6) reduces to

$$\frac{dC_S}{dt} = \frac{Q_p}{V}C_0^S \tag{7}$$

Substituting Eq. (2) into Eq. (7) gives

$$\frac{dC^S}{dt} = \frac{C_0^S}{V}\frac{1}{\sqrt{b^2 + 4at}} \tag{8}$$

Integrating Eq. (8) results in

$$t = a(VC^*)^2 + bVC^*$$

or

$$t = \frac{K_1}{2}\left[\frac{VC^*}{A}\right]^2 + k_2\left[\frac{VC^*}{A}\right] \tag{9}$$

where

$$a = \frac{K_1}{2A^2}, \quad b = \frac{K_2}{A}, \quad C^* = \left(\frac{C_f^S}{C_0^S} - 1\right) \tag{10}$$

Mass Balance Equation for Sodium

The wash stage involves the washing of sodium and the mass balance equation for the quantity of sodium in the system results is presented as

$$V\frac{dC^{Na}}{dt} + Q_w C^{Na} = 0 \tag{11}$$

where C^{Na} is the bulk phase concentration of sodium and Q_w is the permeate rate associated with wash.

The permeate rate associated with washing is given (17) as

$$Q_w = \frac{1}{2aV_f + b} \tag{12}$$

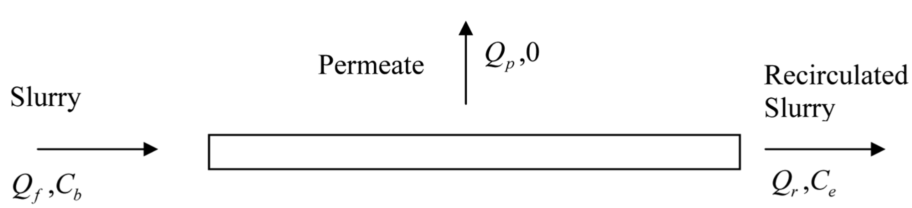

FIG. 4. Mass and flow balances for membrane tube.

where V_f is the final volume of permeate during dewatering and equal to C^*V. Q_w can also be presented as

$$Q_w = \frac{1}{\frac{K_1 V C^*}{A^2} + \frac{K_2}{A}} = \frac{A^2}{K_1 V C^* + K_2 A} \tag{13}$$

Solving Eq. (11) results in

$$C^{**} V = Q_w t_w \tag{14}$$

where $C^{**} = \ln(C_f^{Na}) - \ln(C_0^{Na})$. Substituting Eq. (13) into (14) results in

$$t_w = \frac{K_1 V^2 C^* C^{**}}{A^2} + \frac{K_2 C^{**} V}{A} \tag{15}$$

Also, a cycle time can be derived from

$$t_{cyc} = t_d + 2t_w + t_{etc} \tag{16}$$

where t_{cyc} is the cycle time, t_d is the dewatering time, t_W is the wash time, and t_{etc} is the time to leach, clean and transfer.

Incorporating Eqs. (9) and (15) into Eq. (16) results in

$$t_{cyc} = K_1 \left(\frac{V}{A}\right)^2 C^*(C^* + 2C^{**}) + K_2 \frac{V}{A}(C^* + 2C^{**}) + K_3 \tag{17}$$

where K_1 and K_2 have previously been defined and K_3 is a constant associated with leaching, cleaning, and transfer. The values for K_1 and K_2 were derived above and $K_3(t_{cyc})$ is taken as 68 hours.

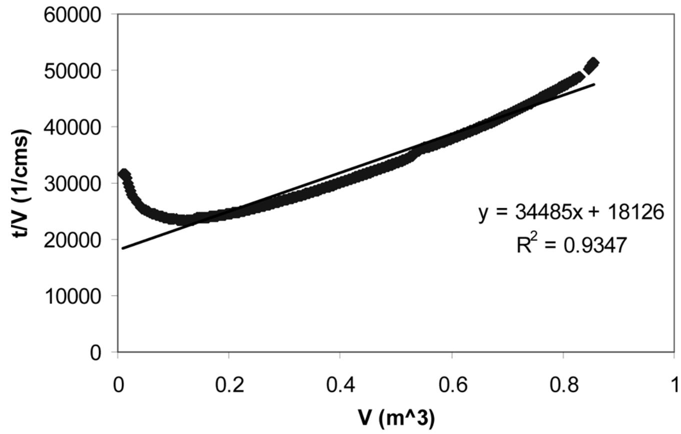

FIG. 5. V vs. t/V curve.

RESULTS

Given that models are available for permeate rates and mass balance equations for bulk phase solids and sodium and that these models are applicable to the UFP at Hanford, then design curves can be developed to determine the minimum membrane surface area for a given volume of slurry to treat and the prescribed time. Other constraints included a particular C^*, C^{**}, and $\{K_1, K_2\}$.

The values for $\{K_1, K_2\}$ were taken from an appropriate empirical study that are results of a bench-scale or pilot-scale study. The empirical study utilized in this work is Duignan (10) and the conditions for the best permeate flux rates were modeled to determine {a, b}, see Fig. 5. The results of a minimum membrane surface area analysis for dewatering times appear in Fig. 6. The conditions given in Fig. 6 are for three ratios of bulk phase concentration, a volume of slurry at 82.5 m^3, and {a, b} = {576 min/m^6, 302 min/m^3}. The same set of conditions were

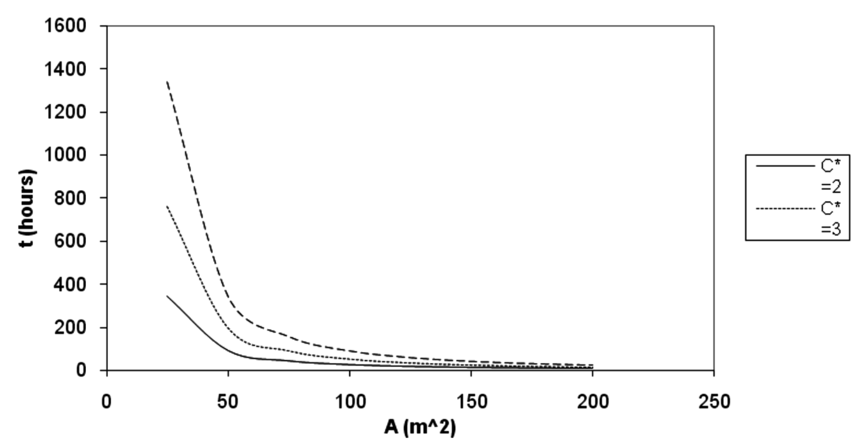

FIG. 6. Design curves for dewatering step.

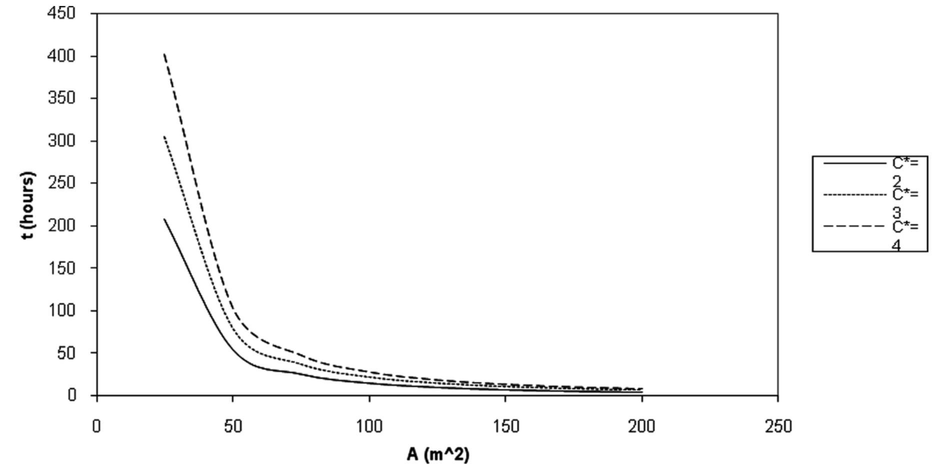

FIG. 7. Design curves for wash step.

utilized below in Figs. 7 and 8 for the development of design curves for the wash and cycle times.

DISCUSSION

The use of a dead-end filtration model for cross-flow filtration is presented in Fig. 5. It is apparent from this figure that the cake layer quickly develops, there is little compression, and along with no significant mass accumulation on the membrane (11) Eqs. (2) and (7) are appropriate. An upcoming study will show that cross-flow filtration systems can behave as dead-end filtration when

1. the membrane permeability is low or
2. surface forces are greater than shear forces in terms of cake material re-suspension into the bulk phase.

Currently, it has been found that several cross-flow systems treating disparate materials (radioactive waste, acai juice, bentonite + water solution) act dead-end, and the upcoming work will establish why this is and delineate how often cross-flow systems behave as dead-end filtration.

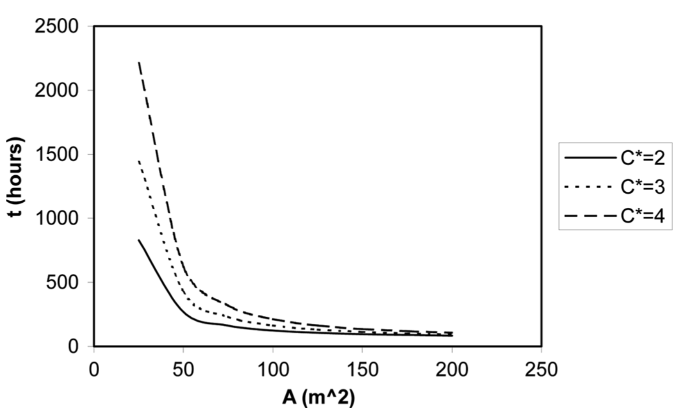

FIG. 8. Design curves for cycle times.

A simple dynamic model for bulk phase concentration of solids is available for the UFP and represented by Eq. (7). Using this model and a model for permeate rates associated with dewatering operations, an equation was derived for the minimum membrane surface area to for a given dewatering operation (Eq. (9)). A design curve associated with this equation is given as Fig. 6.

As can be seen in Fig. 6, a quadratic relationship exists between the membrane surface area and the dewatering times. Similar relationships exist between the volume to treat (or C^*) and the dewatering times.

An important concern of the production maximization program is to properly address issues of scale between a bench (pilot) scale study and a full-scale process. This work has addressed many of these concerns in terms of scaling from one membrane surface area to another, one volume to treat to another, or one ratio of bulk phase concentrations to another. Other issues of scale that may include morphological considerations of the membrane and cake are beyond the scope of this study.

For wash operations, a mass balance equation was derived for the sodium within the system and a prescribed model for permeate rates was derived from Geankoplis (17). Associated with the derived equation for wash times is a design curve where for a given wash volume, C^{**}, t_w, the minimum membrane surface area can be determined. As shown in Fig. 7, there is a quadratic relationship between membrane surface areas and wash times.

A third design curve was also developed for cycle times and is given as Fig. 8. The same quadratic relationship is seen in Fig. 8 as in Figs. 6 and 7. It should be noted that from Figs. 6–8 large differences exist from one C^* to another C^* below $50\,m^2$ and above $200\,m^2$ there is essentially no difference. Also, above $200\,m^2$ increases in the membrane surface area do not appear to give an appreciable decrease in treatment time. This would lead to the observation that the best operation condition lies between $50\,m^2$ and $200\,m^2$, which would have to be supported with further research.

One area that needs to be addressed as future research is that the permeate rates associated with the wash step are in fact not constant but have been shown to increase with time. No apparent reason is given for these results. One possible explanation could be that as the salts concentration decreases with time the viscosity of the permeate also decreases with time. This decreasing permeate viscosity results in an increase in permeate rates.

Another area to explore is empirical validation of these design curves from one scale to another. Such a consideration was beyond the scope of the present study. The reason is that many studies have been conducted on the proposed UFP at Hanford but the results of these studies often disagree from study to study. Often, this disagreement is because issues of scale have not been properly addressed between the bench (pilot) scale apparatus and a full-scale process, but also the developed cakes can differ from one experiment to another that utilizes the same simulant, conditions, and membrane. In one instance, Duignan (10), the only variation was how the simulant was prepared in terms of mechanical mixing, but different values for {a, b} are exhibited.

CONCLUSIONS

A theoretical study was conducted to derive equations for dewatering, washing, and cycle times associated with a diafiltration model ultrafiltration applied to radioactive waste. The derived models have addressed issues of scale between a bench-scale (or pilot-scale) and a full-scale process. Design curves for minimum membrane surface areas associated with a given dewatering, wash, or filtration operation were presented in this paper. These results are based on mass balance equations for solids and sodium within the system and permeate rates associated with dewatering and wash operations. This work has addressed differing membrane surface areas, and the volume to treat or range bulk phase concentrations during dewatering. Issues of scale need to be addressed when scaling bench (pilot) scale results to a larger scale.

The results in these design curves are likely conservative because the permeate rates associated with the wash operations may not be constant and could in fact increase with time. Essentially, the results of a bench-scale dewatering study can be utilized to find {a, b} associated with t/V vs. V and this would form the basis to design curves similar to those presented in this paper. Once these design curves are developed the minimum membrane surface areas are known for a given volume to treat, the ratio of bulk phase concentrations (difference of logarithmic concentrations of salts), and the prescribed dewatering time, wash time, or cycle time.

NOTATION

{a, b}	parameters within linear model for t/V versus V
A	membrane surfaced area
C^S	bulk phase concentration of solids
C^{Na}	bulk phase concentration of sodium
C_e	concentration of solids in return
C_f^*	final bulk phase concentration of solids or sodium
C_0^*	initial bulk phase concentration of solids or sodium
C_s	concentration of solids within the developing cake layer
K_1, K_2	parameters associated with dewatering
K_3	parameter associated with leaching, cleaning, and transfer time
Q_f	flow rate from holding tank
Q_r	flow rate in return
Q_p	permeate rate
t_{cyc}	cycle time
t_d	dewatering time
t_w	wash time
V	volume of slurry to treat
V_f	volume of permeate collected during dewatering step
V_p	permeate volume
α	measure of the compressibility of the cake
Δp	transmembrane pressure differential
η	feed rate into UFP
ρ_{super}	density of supernatant
ρ_s	density of solid
μ	viscosity of permeate

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