Modeling Pilot-Scale Crossflow Filtration of Simulated Nuclear Waste

J. A. Gantt, E. P. Gatzke, V. Van Brunt,

R. B. Haggard, T. J. Deal, C. C. Stork (USC),
& M. R. Poirier, S. D. Fink (SRTC)

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Crossflow Filtration Motivation

- Significant amounts of US radioactive waste slated for disposal
  - 52,000 tons of spent fuel
  - 91 million gallons of liquid high-level nuclear waste
- Savannah River Site currently is disposing of 34 million gallons of high level waste.
- The Defense Waste Processing Facility (DWPF) is processing this high-level waste to encapsulate the radionuclides in borosilicate glass.
- The DWPF needed to separate the high-level waste from low-level waste in a manner that is safe, large-scale, has limited human interaction and is able to handle dense suspensions well.
- Separation processes needed to remove cesium-137, strontium-90, and select actinides
  - Monosodium titanate sorption of strontium-90 and select actinides followed by crossflow filtration is first step
  - Filtrate will be treated by solvent extraction to remove cesium-137

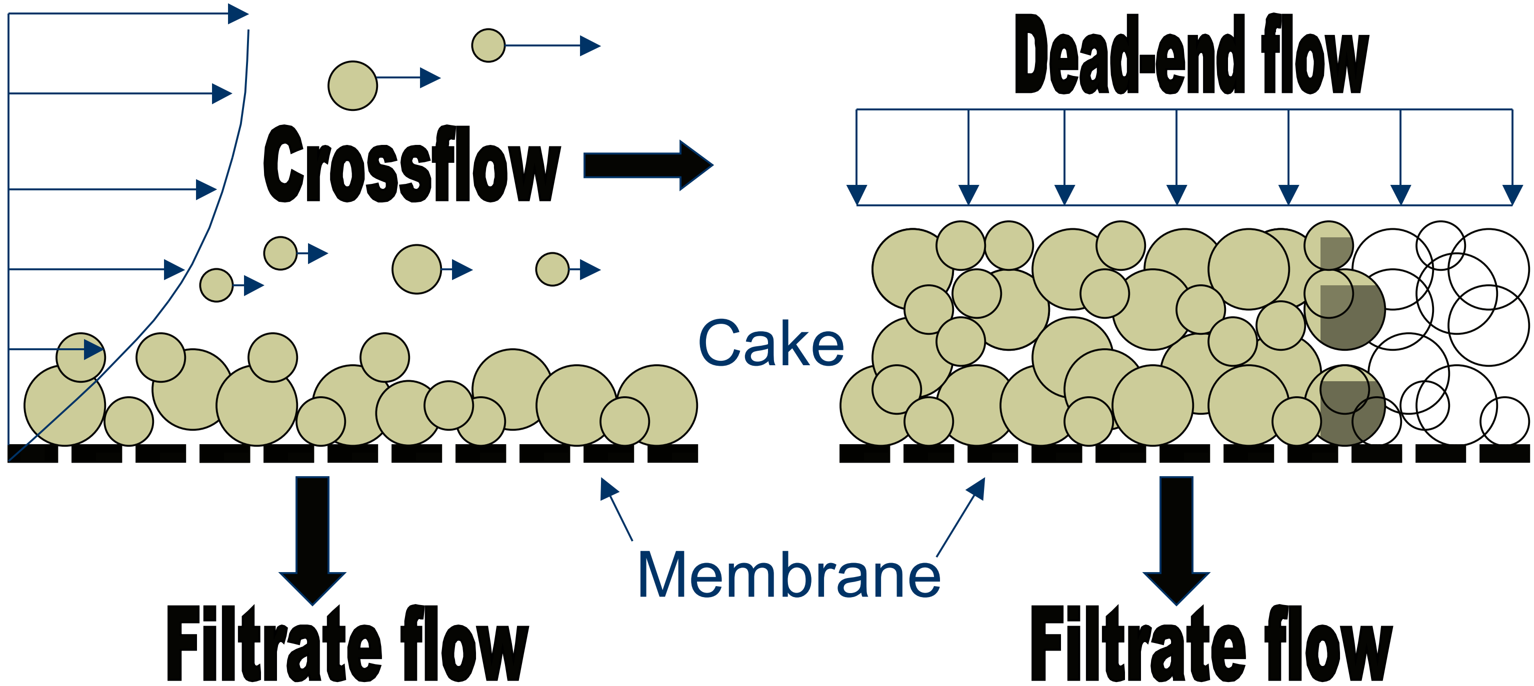
Introduction

- Filtration is a basic unit operation for the removal of undissolved solids from liquids. There are two basic categories:

Crossflow filtration	***Traditional filtration***
Long periods of separation	Frequent cleaning and stopping of process
Minimal human contact	Cleaning requires frequent human contact
Filtration of dense and well dispersed suspensions	Most efficiently filters well dispersed suspensions

- Because of its characteristics, crossflow filtration is ideally suited to processing nuclear waste.

Crossflow Filtration Versus Traditional Filtration

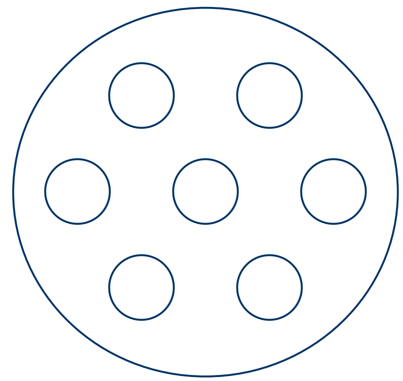


Filtration Research Engineering Demonstration **(FRED)**

- The Filtration Research Engineering Demonstration (FRED) is a crossflow filtration pilot plant at the University of South Carolina.
- Non-radioactive simulants of nuclear waste are filtered to determine filtration process characteristics in long-term operation.
- FRED was designed with the SRS to verify operational aspects of the Late Wash and ITP facilities, and is now used with the Defense Waste Processing Facility located at SRS.
- Currently used to perform pilot-scale testing for the development of new waste treatment facilities

Filters at FRED

Mott 0.5 micron filter



Arranged like shell and tube heat exchanger
7 filters in a tube with a ¾” o.d.; 5/8” i.d.; 0.5 mm pores (nominal)
10 ft length
same size tubes as have been used in facilities at SRS

Cake Removal

Filter can be cleaned without human contact.

There are three methods of filter cleaning and cake removal:

- **Backpulse** - Strong reverse pressure wave used for for membrane cleaning.
  - Backpulsing is an important procedure to maintain operation at FRED
  - Resulting dynamics are difficult to predict
- **Scour** – Method of filter cleaning where filtrate flow is shut off and axial velocity is increased to break down cake formation.
- **Chemical Clean** – Chemical procedure where oxalic acid followed by sodium hydroxide are added to the entire system (filter, piping, tanks, valves, etc.) before a test is performed to clean all impurities.

Dynamic Modeling

[redacted]: Create a dynamic model for FRED based on pilot-scale process data

- Constant Process Variables:
  - Concentration
  - Transmembrane pressure
  - Axial velocity
- Dynamic Process Variables:
  - Number of backpulses
  - Filtrate flux
  - Time of filtration

Crossflow Filtration Setup at FRED

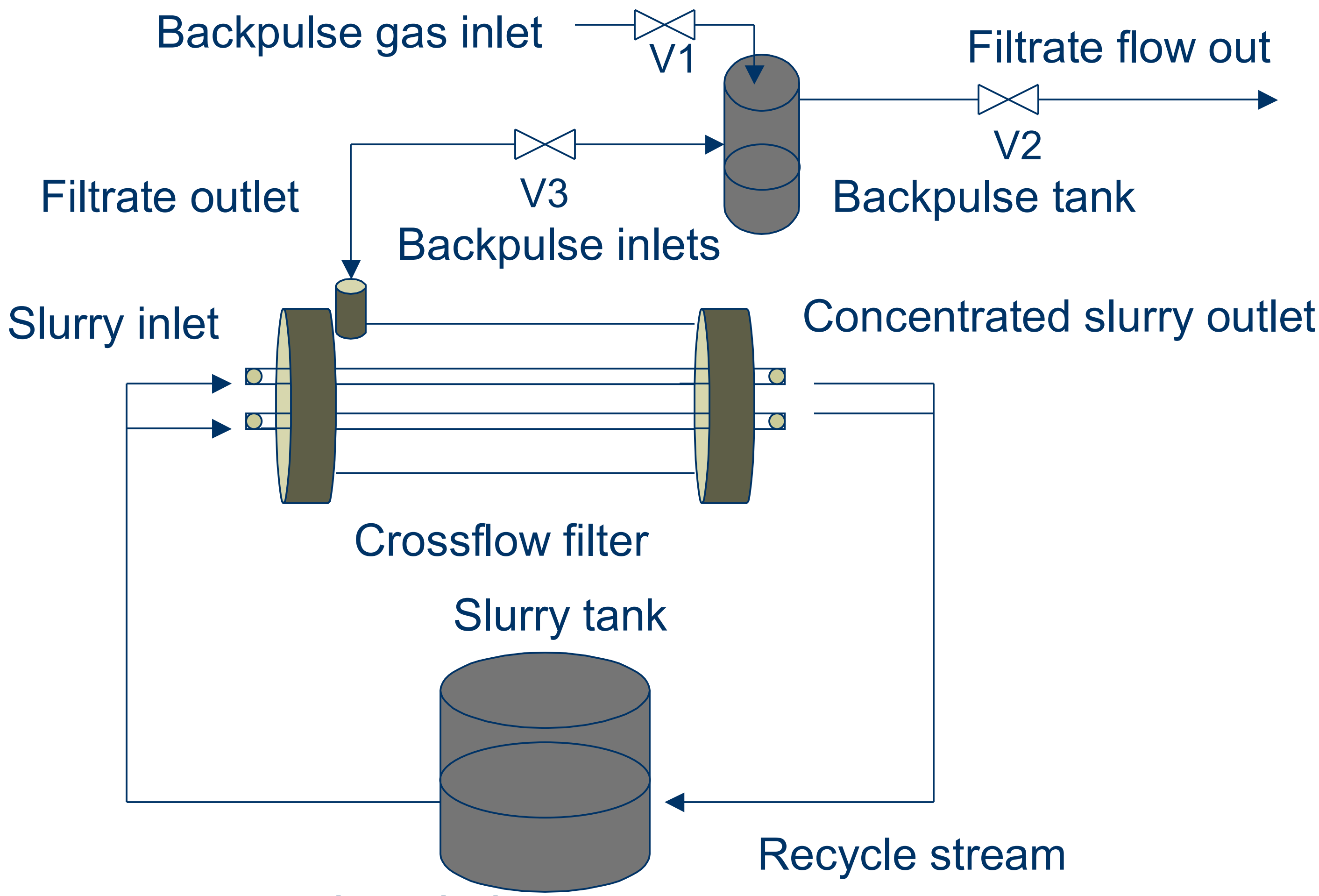


Figure 1: FRED process description

Crossflow Filtration Theory

- Darcy's Law (Coulson and Richardson(1968)):
  - $J = \Delta P / \mu R_{tot}$
- The total resistance term:
  - $R_{tot} = R_{membrane} + R_{cake} + R_{fouling}$
- $R_{membrane}$ is the resistance to flow due to the filter membrane.
- R_{cake} is the resistance to flow due to cake formation.
- $R_{fouling}$ is the resistance to flow due to the fouling of the filter membrane.

Membrane Resistance, $R_{membrane}$

- $R_{membrane}$ is the filtrate flow resistance due to the filter membrane.
- $R_{membrane}$ can be calculated by using a pure water flux through the filter.
- With a viscosity and transmembrane pressure known the membrane resistance can be calculated:
  - $R_{membrane} = 1x10^{7}\ cm^{-1}$

Cake Resistance, R_{cake}

- If the slurry is incompressible, cake resistance can be calculated based on conventional theory (McCarthy et al., 2001)
  - $R_{cake} = \alpha\, m_c / A_t$
  - α is a specific cake resistance per unit weight
  - A_t is the total membrane area
  - m_c is the accumulated mass of slurry through the filter.
- Total mass of the cake is proportional to the total volume of slurry that has passed through the filter after a backpulse:
  - $R_{cake} = f(J_{tot}(t))$
  - J_{tot} is the total volumetric flux of filtrate through the filter at time t after a backpulse was performed.

Cake Resistance, R_{cake}

- Concentration polarization arises in a pressure-driven membrane process when a gel layer of slow moving solids form above the cake.
- The concentration decreases away from the membrane concentration, C_m, until it reaches the bulk concentration, C_b.
- Brusilovsky et al. (1992) define the concentration polarization modulus :
  - $J = k \ln(C_m / C_b)$
  - k is a mass transfer coefficient.
- The rate of concentration polarization layer formation is first order (Murkes and Carlsson (1988)):
  - $J = J_0 \exp(-kt)$

Fouling Resistance, $R_{fouling}$

- Backpulsing does effectively remove filter cake, but there still remains particles in the membrane.
- Backpulsing reported to remove 97.5% of particles from a filter membrane (Sondi and Bhave, 2001).
- An accumulation of particles in the membrane creates a fouling resistance, reducing filtrate flux.
- Furthermore, backpulsing can lodge particles into a filter membrane, contributing to a fouling resistance.
- The fouling resistance is proportional to the number of backpulses performed on a system after a chemical wash:
  - $R_{fouling}=f(N_{BP})$ where N_{BP} is the number of backpulses following a chemical clean.
  - Fouling, therefore, is a cumulative resistance over time.

Experimental Plan

- Many tests have been performed at FRED varying:
  - Chemical species in the slurry (i.e., high iron vs. low iron, high aluminum vs. low aluminum).
  - Operation and cleaning sequences.
  - Filtration conditions (insoluble solids concentration, axial velocity, transmembrane pressure, filtration time)
- The test selected to model was intended to examine the filtration of sludge/ MST suspensions over a range of solid concentrations.
- Test parameters:
  - 4 slurry concentrations from 620 mg/L to 9300 mg/L of solids
  - Axial velocity ranged form 4-20 ft/s.
  - Transmembrane pressure typically ranged from 20-50 psid.

Modeling of Experimental Data

- To find a dynamic model, first define total resistance from Darcy's Law:
  - $\Delta P/\mu J(t) = R_{tot} = R_{mem} + R_{cake} + R_{fouling}$
- The membrane resistance is constant:
  - $R_{mem} = constant = C_1$
- The cake resistance is a linear function of the total filtrate volume passing through the filter since the last backpulse:
  - $R_{cake} = \alpha + \beta\, J_{tot}$ where α and β are then a function of cake formation parameters:
  - α & $\beta = f(C, t)$
- Fouling resistance is a linear function of the number of backpulses :
  - $R_{fouling} = C_2 N_{BP}$

Modeling of Experimental Data

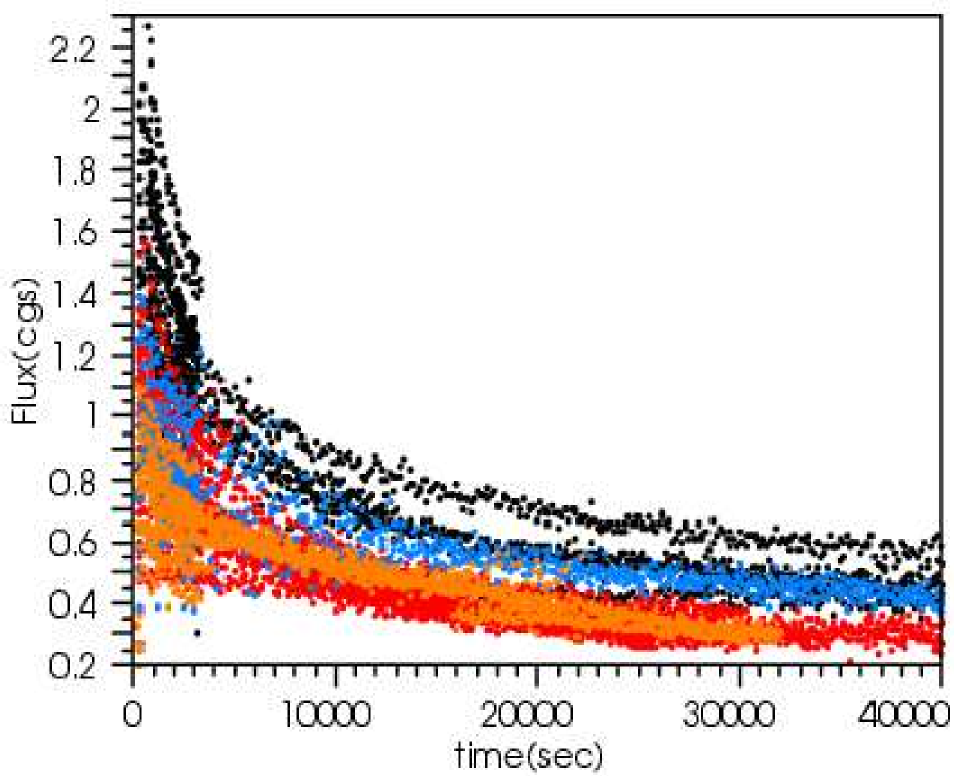


Fig. 2: Typical experimental FRED flux response

- Long and short term process data:
  - 63 short term points chosen (1 hour)
  - 16 long term points (5-15 hours).
- The dynamic model must fit both sets of data so the data was weighted to account for short term point more accurately.

Modeling Using JMP Statistical Program

- JMP (SAS Institute) statistical program was used for linear regression of process data to produce a model of the system.
- The model found to predict the total resistance was:
  - $R_{tot} = 3.0e8 + (-4100 + 1800C - 4360 \ln C)\, J_{tot} - 2.4e8 \exp(-kt) + 3.6e6\, C + 7.2e5\, N_{BP}$
- At $t = 0$, the membrane resistance was found to be:
  - $R_{mem} = 3.0e8 - 2.4e8 = 0.6e8 = C_1$
  - Experimental value = 1.0e7 cm^{-1}
- The resistance model fit the weighted data to an accuracy of 92.6% and fit all the data 92.1% and all coefficients had a t ratio greater than 18.

Modeling Results

- The final flux model using the total resistance term was found to fit an encouraging 90% of all the data (approximately 10,000 data points). It fit the short term points well, 87.5% and the long term points well 90.45%.

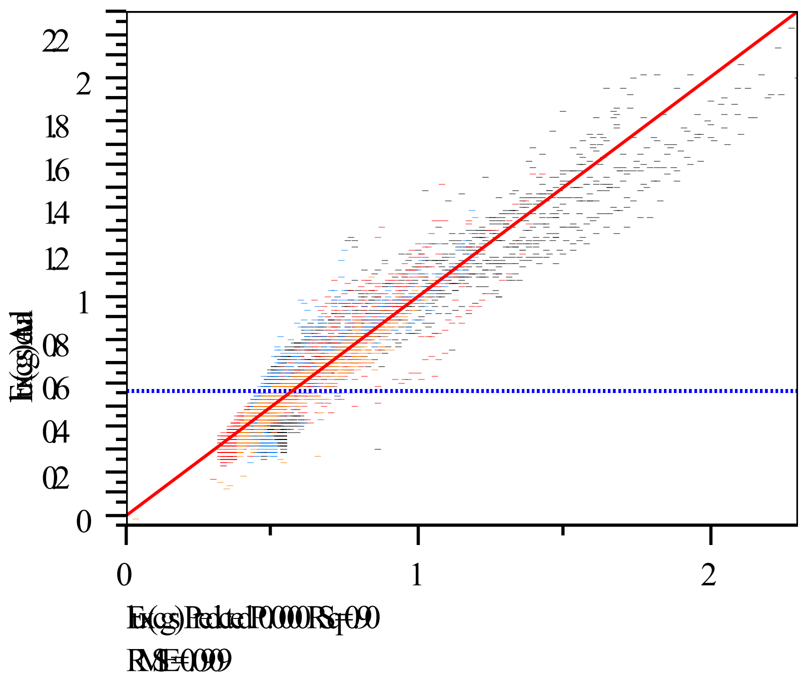


Fig. 3: JMP modeling results using all data.

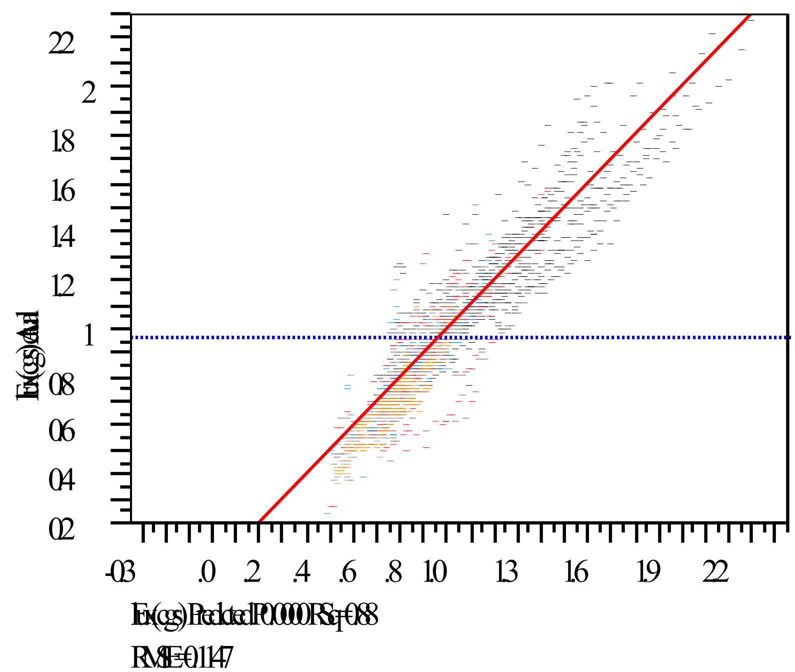


Fig. 4: JMP results using only short term data.

Conclusions

- A model has been developed from crossflow filtration fundamentals that fits the FRED filtration process to 90% accuracy.
- This model shows that the number of backpulses has a large effect on the total resistance.
- A membrane resistance was experimentally calculated to be 1.0e7 cm^{-1} and was calculated from model parameters to be 6.0e7 cm^{-1}.
- This model was shown to effectively fit short term data where the dynamics dominate the process as well as long term data where tests were run at steady state conditions.
- The resistance model fit 92.6 % of process data and the final filtrate flux model fit 90% of process data.

Acknowledgments

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