Filtration of Hanford Tank 241-AP-107 Supernatant Samples Obtained at Prototypic Tank Level and Filtered at 16 °C-22424

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ABSTRACT
Filtration with a metallic media grade 5 filter is planned to be used to remove solids from the Hanford tank supernate streams as part of the Tank Side Cesium Removal system upstream from the cesium removal component. This will provide feed for the Low-Activity Waste Facility at the Hanford Waste Treatment and Immobilization Plant. The average bulk supernate temperatures of the majority of double-shell tanks in the Hanford tank farms currently average around 16 °C during the spring and winter months. Filtration testing was performed with supernatant collected from Hanford tank 241-AP-107 at pump suction height and filtered at 16 °C to assess the impact of this temperature on operations. The testing results demonstrated significantly faster filter fouling than was seen in prior testing that used a sample collected higher in the tank and tested at a higher temperature (25 °C). The solids that were present in the current sample caused significant filter depth fouling that could not be alleviated with a simple backpulse. Additional effort was required to clean the filter by dissolving these solids with recirculating 0.1 M NaOH. Post cleaning, the filter resistance was effectively restored to initial conditions. However, resumed processing of the 241-AP-107 feed at 16 °C continued to result in an increased rate of filter resistance over the same time frame.

INTRODUCTION
The U.S. Department of Energy's Hanford Site houses 56 million gallons (212 million liters) of high-level radioactive waste generated from plutonium production from 1944 to 1988 [1]. The supernatant waste, currently stored in underground tanks, is intended to be vitrified following filtration and ^{137}Cs removal at the Hanford Waste Treatment and Immobilization Plant (WTP) Pretreatment Facility. Because the Pretreatment Facility will not be operational for several years, ^{137}Cs will be removed from low-activity waste (LAW) vitrification feeds using the Tank Side Cesium Removal (TSCR) system in a technology demonstration that will filter and remove cesium from tank waste supernate to support transferring the TSCR-processed waste directly to the WTP LAW Facility. The TSCR system is skid-mounted and employs two key technologies: (1) dead-end filtration for solids removal and (2) ion exchange (IX) for cesium removal. Filtration is necessary to protect the functionality of the IX columns.

A small-scale test platform was established in 2017 to demonstrate these processes in the Pacific Northwest National Laboratory (PNNL) 325 Building, also known as the Radiochemical Processing Laboratory (RPL).

The presence of solids has been observed in previous filtration experiments using waste from Hanford tanks 241-AP-105 and 241-AP-107 [2,3,4]. The first test with 241-AP-107 (AP-107) was performed with samples that were retrieved from the tank within a few weeks of raw water addition (and tank recirculation); solids were observed in those filtration experiments. Additional AP-107 samples were taken 14 months later and filtration showed no recoverable solids [5], indicating that settle-then-decant works to reduce solids in supernatant. Both sets of these samples were taken from relatively high in tank AP-107; however, the pump suction is located much lower in the tank. In addition, the prior tests were performed at the ambient temperature of the hot cell (25 °C), while the TSCR operating temperature may be as low as 16 °C.

Solids obtained from the first sampling event in AP-107 are suspected to be due to tank mixing and formation of precipitates when combined with process water to achieve a dilution from 8.5 to 5.6 M Na for prototypic processing conditions. Formation of solids upon dilution with process water is supported by Daniel et al. [6], who observed that precipitates formed upon dilution of 241-AP-105 simulant with process water. However, reducing the temperature to 16 °C is also suspected of having the potential to produce additional solids.

Thus, the objective of the current work was to perform filtration under prototypic conditions using Mott Grade 5 sintered metal at the targeted flux of 0.065 gpm/ft^2 (2.65 lpm/m^2) planned for TSCR, at the TSCR operation temperature, on feed obtained closer to the pump suction to assess whether these two factors (depth of sample collection in the waste tank and filtration temperature) result in higher solids loading to the filters, and thus lower filter fluxes. Filter resistance as a function of volume of feed filtered was measured and the filter was backpulsed if the differential pressure increased to 2 psi (13.8 kPa) across the filter. Backpulse solutions were collected and solids were found and assessed by microscopy methods.

EXPERIMENTAL

Test Conditions

In November 2020, Washington River Protection Solutions collected 36 supernatant samples (~250 mL each) from tank 241-AP-107 in two batches, at a level near the pump suction (3 ft (0.914 m) from the bottom of the tank) and provided them to PNNL. Two of the sample containers were held for IX batch contacts, leaving 34 bottles for filtration. At the RPL, PNNL composited the 34 remaining samples into 1.5-L polyethylene bottles and chilled the bottles (16 °C setpoint) for approximately 1 week. Filtration testing of the tank waste began on February 9, 2021, using a new Mott Model 6610 (Media Grade 5) line filter with porous end cap. This is a sintered 316L stainless steel filter with a 0.317-in. (8.05E-3 m) porous diameter, 1.463-in. (3.72E-2 m) porous length, and 1.51-in^2 (9.74E-4 m^2) filter area.

Backpulse Dead-End Filter System Description

The filtration system is the same system that was used in the previous year [4] with a few modifications to support reduced-temperature filtration. Modifications included two additional heat exchangers installed in the hot cell (connected to two chillers) to control the temperature of the feed before and during filtration. The first new heat exchanger (trough heat exchanger) kept all the feed at the setpoint temperature until it was added to the backpulse dead-end filter (BDEF) system. The trough heat exchanger has dimensions of 9 in. × 25 in. × 12 in. (width, length, height) (0.23 m x 0.64 m x 0.30 m) with a removable cover on the top. The feed bottles were stored in the trough heat exchanger with a cover until the feed was transferred to the BDEF system.

Once the feed was added to the BDEF, the existing heat exchanger kept the feed at the setpoint temperature in the reservoir and in the BDEF recirculation loop. The second new heat exchanger (clamshell heat exchanger) completely enclosed the filter and associated tubing to keep the feed at the setpoint temperature as it exited the recirculation loop until it was filtered. After filtration, the temperature was no longer controlled.

A piping and instrumentation diagram is provided in Fig. 2**Error! Reference source not found.**. A photograph of the BDEF system installed in the RPL Shielded Analytical Laboratory hot cell is shown in Fig. 1.

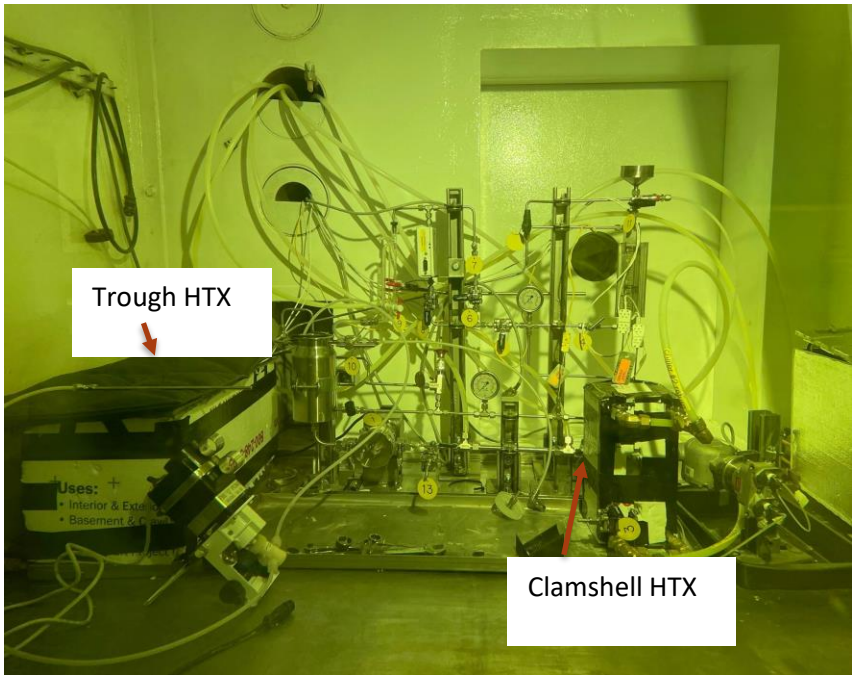


Fig. 1. BDEF system installed in hot cell. HTX = heat exchanger.

The BDEF system is composed of a slurry recirculation loop, a filter assembly, and a permeate system. The main recirculation loop consists of a 1-liter stainless steel container (Eagle, EPV1A), a low-shear quaternary diaphragm pump (Quattro Flow QF150), a heat exchanger, and a throttle valve. The pump speed is controlled by a variable frequency drive that is located outside the hot cell. The slurry flow rate and pressure are controlled by adjusting the pump variable frequency drive (pump speed control) and throttle valve. The recirculation loop provides mixed, pressurized feed to the filter assembly. During the testing described in this report, the slurry temperature was controlled at a 16 °C setpoint.

The filter assembly receives pressurized slurry from the slurry recirculation loop. The filter assembly is composed of a filter, a Rosemount differential pressure transducer, and a flush valve (V3 in Fig. 2). The flush valve is actuated during backpulse operations used to clear solids off the filter and out of the system.

The permeate system receives permeate produced by the filter assembly. The permeate flow rate is controlled with a mass flow controller (MFC), which can control feed in the range of 0.15 to 0.33 liter/hour. (These rates equate to allowable filter areas of 1.5 to 3.3 in^2 (9.7E-4 to 2.1E-3 m^2) assuming flux of 0.065 gpm/ft^2 (2.65 lpm/m^2).) The MFC measures flow rate and density of the permeate while a glass flowmeter is provided as a secondary flow rate measurement device. The permeate system can also perform a backpulse function. Pressurized air can be introduced (V12 in Fig. 2) into the backpulse chamber and used to force permeate (or other fluids) backward through the filter and out of the system through V3.

The Mott 6610 filter used in testing is cylindrical, with dimensions of 0.317-in. (8.05E-3 m) diameter × 1.5-in. (3.81E-2 m) length and a filtration area of 1.51 in^2 (9.74E-4 m^2). The filter element is fabricated from a seamless sintered stainless steel tube that is a closed/dead-end porous tube (with a porous end cap); the open end is welded to a pipe-reducing bushing. At 0.065 gpm/ft^2 (2.65 lpm/m^2), the rate of filter processing is 3.7 L feed per 24-hour day. Fig. 3 shows a schematic of the filter assembly and a photo of the filter.

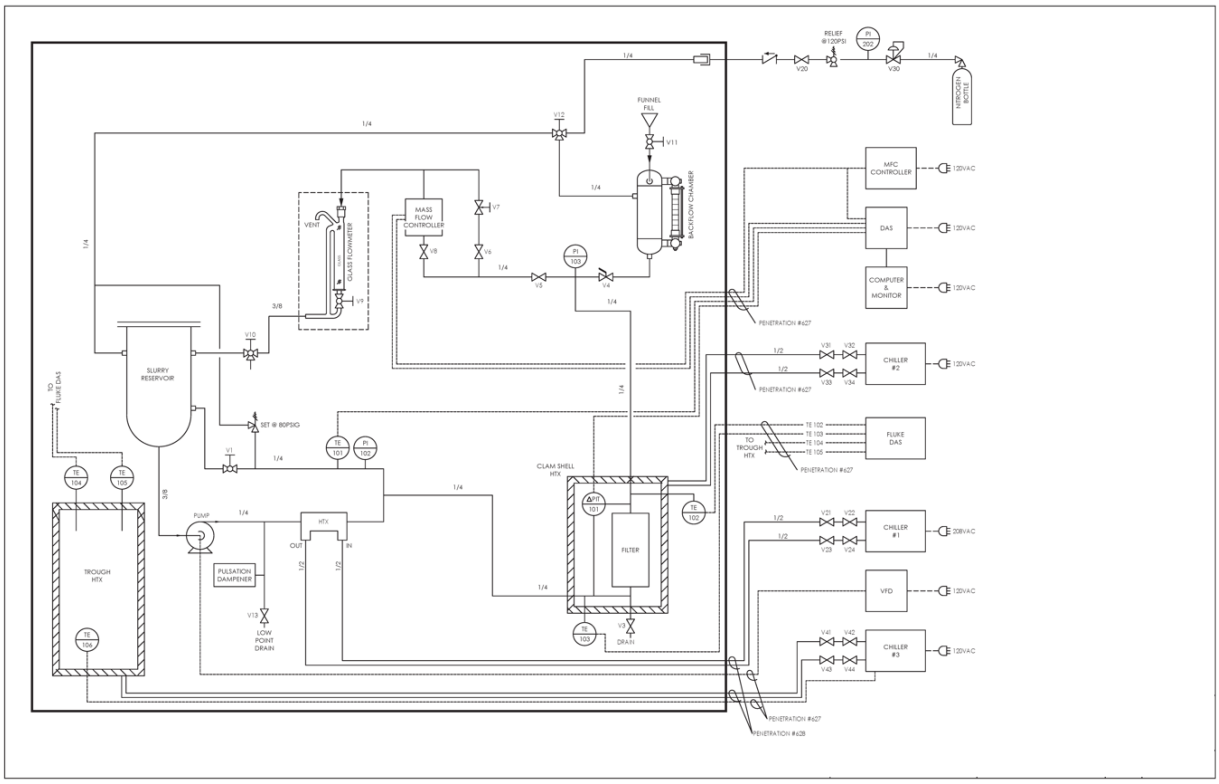
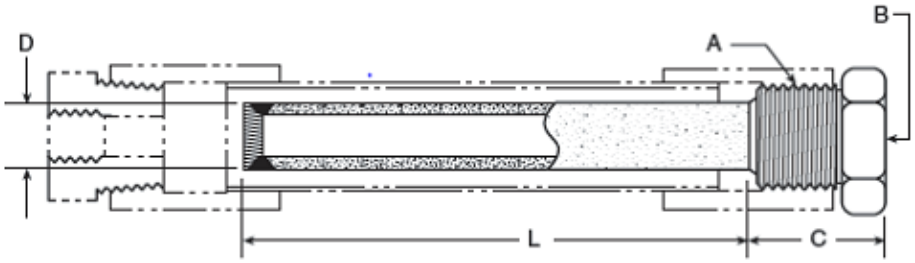


Fig. 2. BDEF piping and instrumentation diagram.



(a) (b)

Fig. 3. (a) Filter housing schematic[1] (note that the 6610 series filter was welded to a 3/8-in. (0.01 m) pipe fitting, making the configuration similar to the 6480 series illustrated here); (b) photo of modified filters with filter housings removed.

[1] Mott 6480 line filter from https://mottcorp.com

System Operation during Testing

The evolutions used to test the diluted AP-107 waste samples are outlined below.

1. Clean water flux (CWF) measurement: The CWF measurement serves as a system leak test and provides a baseline measurement of the filter resistance; it was conducted with 0.01 M NaOH solution at nominal test conditions (2.57 mL/min) and operated for 16 minutes. After the CWF, the BDEF system was drained.
2. Filtration of AP-107 – filtered AP-107 feed with the BDEF at nominally 2.57 mL/min: The targeted filtration rate is based on scaled flux used during AVANTech testing.[2] The filtration rate was controlled with an MFC. Permeate was sampled after approximately 1/3, 2/3, and 3/3 of the feed had been filtered.
3. The filter was backpulsed when the differential pressure was greater than 2 psid (13.8 kPa). Each backpulse used a charge of pressurized air to push nominally 26 mL of filtered permeate through the filter at a rate of nominally[3] 1.5 mL/s.
4. The filter was cleaned after approximately 39 hours of filtration operations (and nine periodic backpulses) because of the marked reduction in permeate recovered by filter backpulse. The filter was cleaned after draining the AP-107 from the BDEF system. (Drained AP-107 was stored in the trough heat exchanger and maintained at the 16 °C setpoint temperature.) A solution of 0.1 M NaOH was added to the BDEF system and pumped through the filter. The recirculation pump was turned off and the filter soaked in the hydroxide solution for 2 hours without temperature control. A measurable decrease in filter resistance during this recirculation indicated that the 0.1 M NaOH was likely dissolving some of the solids that had deposited on the filter. After the filter soak, the filter was backpulsed. Then, the BDEF system was drained of the cleaning solution and filtration of AP-107 continued (second cycle). The filtration test concluded after approximately 14 additional hours of filtration operations, during which time an additional four backpulses were conducted to address increases in transmembrane pressure.
5. The BDEF system was drained.
6. Filter cleaning: 0.1 M NaOH was added to the BDEF system and pumped through the filter. The recirculation pump was turned off and the filter soaked in the hydroxide solution for 2 hours. After the filter soak, the filter was backpulsed, then the BDEF system was drained of the cleaning solution.
7. The BDEF system was rinsed using 0.01 M NaOH and subsequently drained.
8. Final CWF measurement: After rinsing, another CWF test was conducted using fresh 0.01 M NaOH solution using the same conditions outlined in Evolution 1.
9. The BDEF was laid-up for post-test storage.

TABLE I provides a mass balance for the BDEF testing. A total of 11,363 g of AP-107 supernatant was added to the BDEF system during testing, and a total of 11,175 g was removed. The missing mass (~190 g) is likely due to evaporation and material that wets the inside of the BDEF system, is not recoverable, and represents less than 2% of the initial feed.

[2] 0.306 gpm (1.16 lpm) through 4.7 ft^2 (4.37E-1 m^2) of Mott sintered metal filter (0.065 gpm/ft^2 (2.65 lpm/m^2)) obtained from *TSCR Dead End Filter Scoping Test Summary*, presentation by AVANTech Inc., November 13, 2018, Richland, Washington.

[3] Scaled from 2.2 gpm/ft^2 (89.6 lpm/m^2) per AVANTech communications. Tracy Barker (AVANTech VP and principal engineer), "RE: TSCR Filter Cleaning Procedure," email message to Jarrod Allred (PNNL engineer), March 20, 2020.

TABLE I. Mass balance – BDEF.

Description	In (g)	Out (g)
Decanted supernate filtration	11,363.1	
Product to IX		9,941.0
Permeate samples		17.5
Backpulse samples		907.5
Drained from BDEF		309.4
Total	11,363.1	11,175.4

The permeate density of the filtered AP-107 product (to be used for IX) was determined gravimetrically by measuring 10 mL from each permeate collection bottle into a volumetric flask and weighing on an analytical balance. The density average was 1.271 g/mL (0.38% relative standard deviation). Results are provided in TABLE II.

TABLE II. Density of filtered AP-107 for Cs ion exchange.

Bottle ID	Vol Flask Tare (g)	Vol Flask Gross (g)	Measured Density (g/mL)
IX-AP7-1	15.0431	27.7491	1.2706
IX-AP7-2	15.1585	27.9176	1.2759
IX-AP7-3	15.2166	27.8959	1.2679
IX-AP7-4	15.1827	27.8597	1.2677
IX-AP7-5	15.1729	27.8050	1.2632
IX-AP7-7	14.4186	27.1903	1.2772
IX-AP7-Last	14.5879	27.2979	1.2710

RESULTS & DISCUSSION

For the duration of the first 24 hours of testing, the filter resistance increased gradually. At 22 hours of operation (3.4 m^3/m^2), there is a discontinuity in the resistance increase. This does not coincide with any of the bottle changes, which has been shown in the past to cause a discontinuity in the resistance increase. However, this type of discontinuity has been seen previously and is generally associated with sloughing of solids from the filter surface. After this event, the filter resistance continued to increase until the 2 psid target was reached after 25 hours and 53 minutes of operation (4.07 m^3/m^2). A backpulse was performed and filtration was resumed. The resistance was significantly reduced by the backpulse but did not return to the original condition ($7.7x10^9$ m^{-1} after the first backpulse versus $4.3x10^9$ m^{-1} at the start of testing).

As seen in Fig. 4, the resistance increased faster after the first backpulse (less volume filtered at a constant flow rate = shorter time interval); it reached the 2-psi (13.8 kPa) threshold after 0.914 m^3/m^2 volume filtered. This behavior was repeated after the second backpulse and reached the pressure limit in significantly less time. After the third backpulse, testing was stopped for a short duration to replace the MFC. Once testing was restarted, the duration between backpulses continued to shorten, as seen in Fig. 5. In an attempt to extend the period between backpulses, the target pressure was allowed to increase to 4 psi. However, this did little to extend the time period as the filter continued to foul ever faster (see TABLE III).

After the ninth backpulse, the backpulse frequency had become untenable as the quantity of backflush fluid used to clear the solids outpaced the permeate production rate. It was decided to perform a more extensive filter cleaning.

As mentioned previously, the filter was cleaned at this point by draining the AP-107 feed into chilled holding containers, introducing 0.1 M NaOH into the feed vessel, and recirculating this solution through the system to ensure the filter was filled with 0.1 M NaOH. During this operation, the filter resistance decreased measurably, indicating that this process alone was likely dissolving some of the solids that had deposited on the filter. The filter was subsequently allowed to soak for 2 hours before another backpulse was performed and filtration restarted after replacing the cleaning solution with AP-107 feed. This cleaning protocol improved filter performance but did not restore the system to the original condition. While the initial post-cleaning resistance (3.9×10^{9} m^{-1}) was effectively the same as the initial condition (4.3×10^{9} m^{-1}), the filter resistance increased much faster after the cleaning than during the initial testing (see Fig. 6). Testing was continued with backpulses performed as required to process the balance of the feed, which was accomplished with four subsequent backpulses without having to resort to an additional filter cleaning.

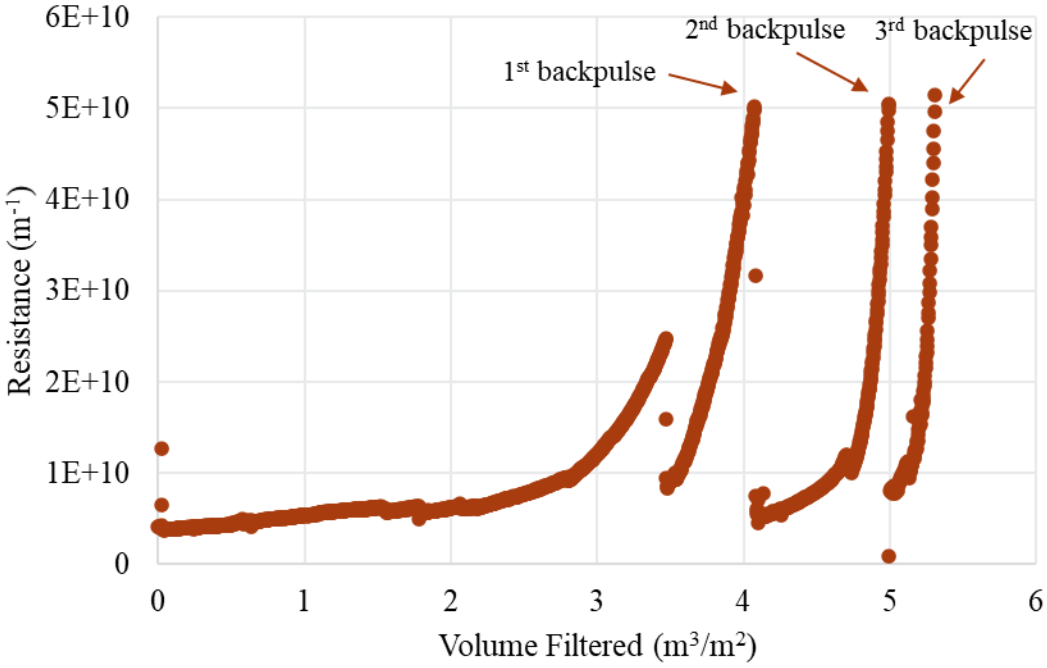


Fig. 4. Filter resistance for filtration prior to cleaning, backpulses 1 through 3.

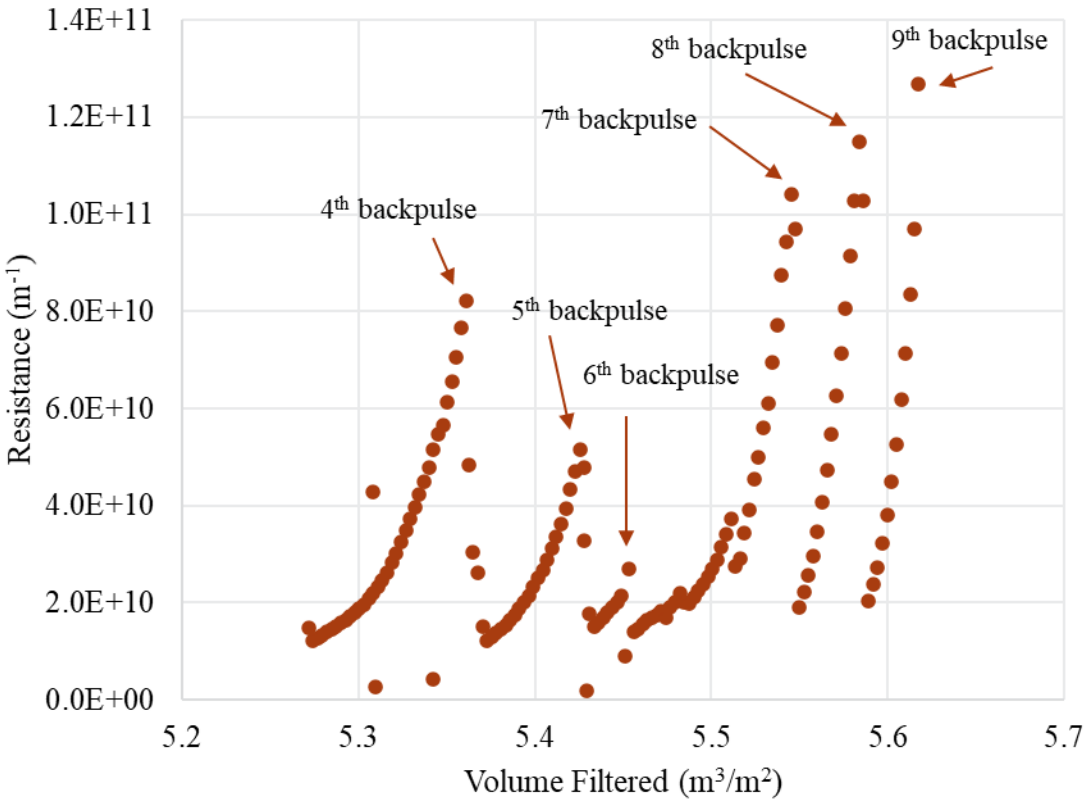


Fig. 5. Filter resistance for filtration prior to cleaning, backpulses 4 through 9.

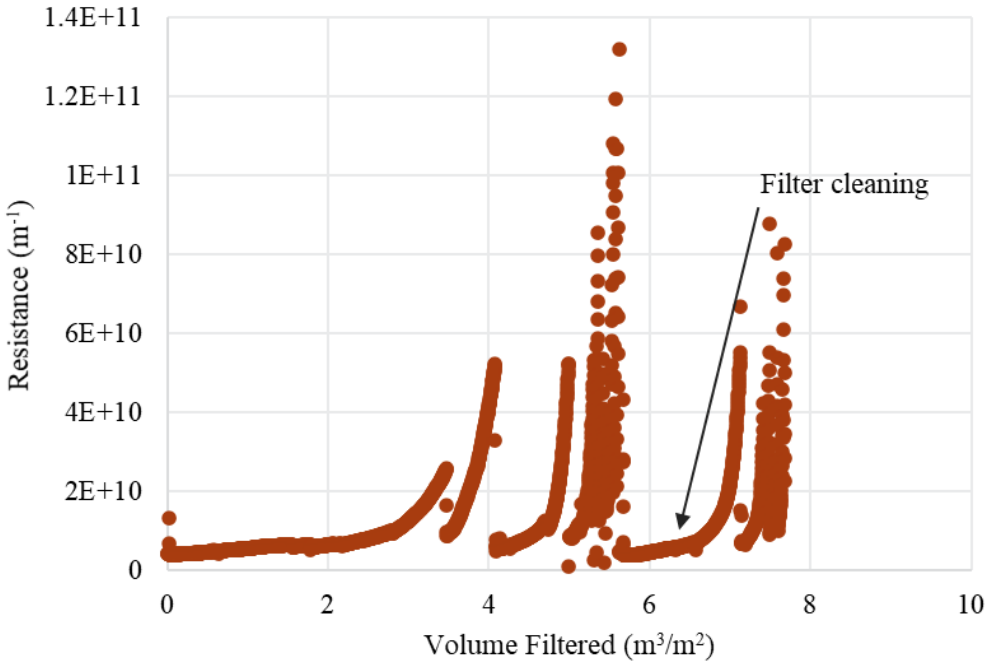


Fig. 6. Filter resistance before and after filter cleaning.

A comparison of the efficacy of backpulsing shows that while the initial resistance was restored significantly by a backpulse, the time to reach the 2-psid action threshold deteriorated quickly as subsequent backpulses were performed. TABLE III provides the test parameters prior to backpulsing, highlighting the reduced volume filtered after each successive backpulse (reduced volume filtered corresponds to shorter time). It was observed that simple backpulsing quickly became ineffective at dislodging particles from the filter surface and subsequent filter fouling accelerated. In contrast, flowing dilute caustic solution through the filter apparently dissolved some of the solids. Note, however, that this dissolution appears far from complete. While the initial resistance appears similar, the duration to reach 2 psi was significantly reduced (by roughly 2/3) after the filter cleaning. These results suggest that some of the solid species may not dissolve in the cleaning solution used for this cleaning protocol.

Note that the effectiveness of the TSCR cleaning protocol is expected to fall somewhere between a backpulse and the cleaning protocol used in this test. The TSCR protocol does not actively flow solution through the system as was done to charge the test platform filter. Thus, the test platform provides a reasonable bound on expected full-scale performance. That is, performance of the TSCR system should neither be worse than observed with the backpulse nor better than observed with the test system cleaning protocol for this type of feed.

The reduced volume filtered after each successive backpulse is consistent with depth fouling of the filter. Depth fouling is characterized as a slow increase in filter resistance as the pores are filled with solids. Once nearly all the pores are filled, the filter resistance begins to increase rapidly, as additional solids are deposited on the surface of the fouled filter. This type of depth fouling is also expected to result in the type of backpulse behavior that was observed. Depth fouling will be less amenable to restoration by backpulsing, as the solids will lodge deep in the pores and be difficult to dislodge. This type of filtration behavior is seen when the size of the particles is such that they are not rejected at the surface of the filter, but rather penetrate the depth of the filter.

TABLE III. Test parameters prior to backpulsing.

Test Event	**Filtration Resistance (m^{-1})**	**Volume Filtered (m^3/m^2)**	**Transmembrane Pressure (psid) / (kPa)**	
Backpulse 1	5.21E+10	4.08	2.01	13.9
Backpulse 2	5.22E+10	0.914	2.00	13.8
Backpulse 3	5.34E+10	0.309	2.06	14.2
Backpulse 4	8.54E+10	0.095	3.29	22.7
Backpulse 5	5.34E+10	0.063	2.06	14.2
Backpulse 6	6.17E+12	0.024	2.20	15.2
Backpulse 7	1.08E+11	0.095	4.16	28.7
Backpulse 8	1.19E+11	0.038	4.60	31.7
Backpulse 9	1.32E+11	0.032	3.31	22.8
Backpulse 10	6.69E+10	1.51	2.45	16.9
Backpulse 11	8.78E+10	0.358	3.51	24.2
Backpulse 12	8.05E+10	0.097	3.03	20.9
Backpulse 13	7.40E+10	0.076	2.80	19.3
Backpulse 14	8.26E+10	0.023	1.80	12.4

It is desirable to compare these filtration results with previous AP-107 BDEF testing. Work done in 2019 was performed at 25 °C at a permeate flow rate of 0.065 gpm/ft^2 (2.65 lpm/m^2) and with a sample gathered higher in the tank, at 50 in. (1.27 m) below the waste surface and 367 in. (9.32 m) from the bottom of the tank [5].

This previous test was also performed using a larger dead-end filter (3.24 in^2 (2.09E-3 m^2) vs. 1.51 in^2 (9.74E-4 m^2)). This resulted in truncated data sets when compared to the current (2021) testing. Fig. 7 shows that the 2021 testing (16 °C data in the figure) resulted in fouling whereas the 2019 testing (25 °C data) did not experience any increase in filter resistance.

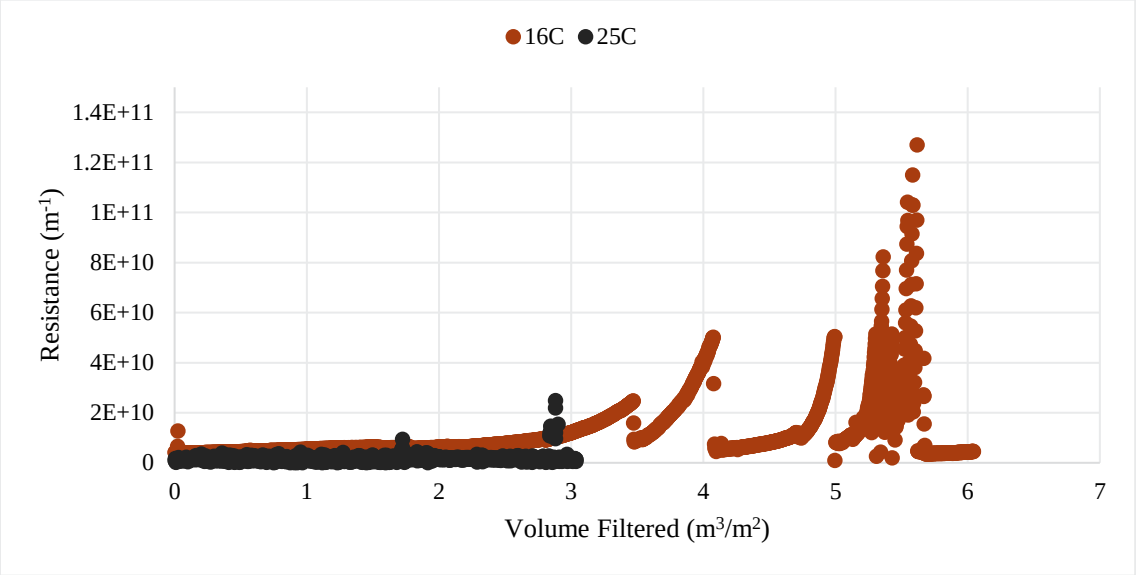


Fig. 7. Comparison of filter data at 25 °C (2019 testing; higher sampling location in tank AP-107) vs. 16 °C (2021 testing; lower sampling location).

CONCLUSIONS

Testing was performed to assess the impact of sampling tank AP-107 at a lower height in the tank and the effect of a lower operating temperature (16 °C) on filter performance. The testing results demonstrated significantly faster filter fouling than was seen in prior testing that used a sample collected higher in the tank and tested at a higher temperature (25 °C). The solids that were present in the current sample caused significant depth fouling that could not be alleviated with a simple backpulse. Additional effort was required to dissolve these solids. Post cleaning, the filter resistance was effectively restored to initial conditions as the initial transmembrane pressure was restored to original levels. However, resumed processing of the AP-107 feed at 16 °C continued to result in an increased rate of filter resistance.

The source of these solids is not fully understood. They may have been associated with the lower sample collection depth or the lower processing temperature. However, the fact that the bulk of the solids readily dissolved with water addition suggests that they are perhaps more likely attributable to formation due to temperature changes. Formation of solids due to temperature change is driven by changes in solubility. Dilution with water would effectively reverse the impact of decreasing temperature. Thus, the result that these solids dissolve with water addition suggests temperature as a primary driver for the solids formation. Therefore, one method to alleviate this type of filter fouling may be to increase the operating temperature of the TSCR filtration system.

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