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INVESTIGATION INTO OPTIMAL CONDITIONS FOR CROSS -FLOW FILTRATION OF HIGH -LEVEL NUCLEAR WASTE

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ABSTRACT

The Savannah River Site has 23 Type III high-level radioactive waste tanks, each with a storage capacity of 1.3 million gallons. These tanks contain nearly 9 million gallons of precipitated salt. To immobilize the waste, the salt is dissolved through water addition, followed by precipitation of the radionuclides through the addition of sodium tetraphenylborate. This precipitate is then concentrated and washed to remove sodium through cross-flow filtration. This waste pretreatment process started radioactive operation in late 1995. During the normal plant operation, the cross-flow filtration system (consisting of two 216-square-foot filter elements) maintains a constant filtrate production rate. This objective is achieved by allowing the operating pressure to increase to maintain a constant filtrate production rate. A maximum pressure differential limit of 40 psig has been imposed on this system. When this maximum is approached, a high-energy backpulse of filtrate removes foulant from the surface of the filter, thereby restoring the filter flux.

This laboratory work examined two key aspects of the anticipated facility operating conditions: the efficacy of using pressure differential to control filtrate production rates and the risk posed to filter performance associated with pore plugging of the filter immediately following the backpulse. Tests used simulated tetraphenylborate precipitate and a bench-scale cross-flow filtration unit consisting of two parallel filter units each 4 feet in length. Tests used slurries containing between 1 and 10 wt % tetraphenylborate to cover the anticipated range of operation. Data collected included both initial flux-decline measurements and steady-state filtrate production measurements. Analysis of these data indicates, for the more dilute slurries, pressure was an effective tool in controlling filtrate flux. However, as the slurry became more concentrated, the ability to manipulate filtrate flux by pressure greatly diminished. Analysis of the initial filtrate decline data using

first- principle models indicates that the primary mechanism for decreasing filter flux involved development of a surface cake. Given the operating constraints of the facility, these results provide guidance for future filtration operation.

INTRODUCTION

The In-Tank Precipitation (ITP) process at the Savannah River Site concentrates cesium tetraphenylborate precipitate to reduce the quantity of high-level radioactive waste processed by the Defense Waste Processing Facility.[1,2] During precipitation, significant amounts of potassium tetraphenylborate form to produce a 1 wt % slurry. The facility concentrates the slurry to 10 wt % solids prior to transferring it to the Defense Waste Processing Facility. This concentration is achieved by cross-flow filtration. These filters typically are operated to maintain a continuous filtrate production rate by increasing the pressure differential as filter performance declines due to fouling. This research program sought to illuminate the nature of this fouling and to provide insights into future operations of this facility.

METHODS AND MATERIALS

This work used a laboratory filtration unit to simulate performance of the full-scale process. Figure 1 contains a sketch of this filtration unit. The experimental equipment can control the operating conditions of each filter individually. A data acquisition system records the axial flow rate (ingpm) and the outlet pressure (in psig) for both filter elements. The filtrate control valve system allows measurement of the filtrate flow rate for each filter either independently or cumulatively. The data acquisition system consists of a Macintosh SE using Workbench Mac software. The data acquisition system records the axial flow rate, the filterflow rate, inlet and outlet pressure, and filtrate pressure. This study used a data acquisition period of 20 seconds. Axial velocities ranged from approximately 1 to 2 m/s (3 to 7 ft/s) over operating pressure differences from approximately 70 to 275 kPa(10 to 40psig). The experiments were performed at 25 •C.

Tests used slurry concentrations ranging from 1 wt % tetraphenylborate solids to approximately 10 wt % solids. These slurries contained 5 molar sodium ion,

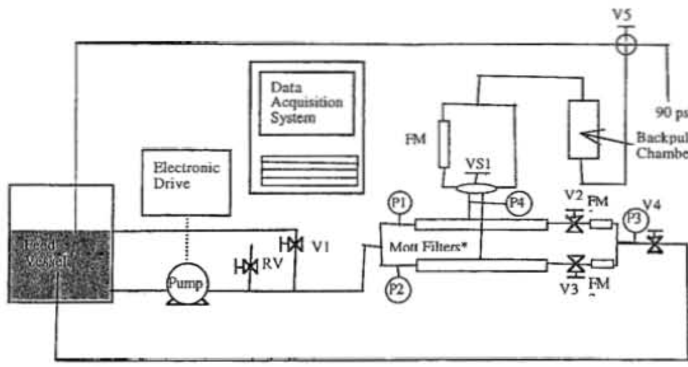

FIGURE 1. Laboratory- scale filtration unit.

approximately 1.5 molar hydroxide ion, 2.0 molar nitrate ion, and 0.5 molar nitrite ion. Slurry concentration was increased by removing 1 liter of filtrate and adding an equivalent volume of 1 wt % slurry to reach the required concentration of tetraphenylborate solids. The tetraphenylborate solids had a mean primary particle size of approximately 1.75 microns (distributed over a range from 0.5 to 10 microns) as determined with a Microtrac particle size analyzer. These particles also form relatively large agglomorates (circa 10 microns). These slurries all had a density of approximately 1.2 g/mL.

At selected solids concentrations, personnel recorded filtrate flux as a function of time at each of four constant operating pressures. Each flux measurement immediately followed a backpulse. These backpulses involved rapidly reversing the filter flow to remove any deposited foulant from the filter surface or from within the filter pores. A decline in filter flux was observed as foulant deposited on the filter at each operating condition of interest.

In addition to laboratory filtration tests, preliminary operations were completed in the In-Tank Precipitation process. These operations used one of the two available 216-ft^2 filter units. (Note that, these elements had the same nominal pore size as those employed in laboratory scale testing). In contrast to laboratory tests, the facility operates to maintain a constant filtrate flow rate by increasing the operating pressure as filter performance degrades. This increase in pressure produces a concomitant decrease in axial velocity through the filter.

THEORY

During filter operation, Darcyís law indicates that filtrate flux (Q) remains proportional to pressure drop (ΔP) across the filter:

$$Q = \frac{\Delta P}{R}, \tag{1}$$

where R gives the resistance of the filter (and any foulant layer). Immediately after a backpulse, the resistance comes from the filter alone. Following the backpulse, solids deposit on the filter, producing fouling. This deposition can occur in one of two ways: solid particles can either deposit in the filter pores or can form a layer on the surface of the filter. When deposition occurs on the surface of the filter, the resistance becomes the sum of the resistance of the filter (R_f) and the resistance of the deposited filter cake (R_c).

Equation 1 then becomes

$$Q = \frac{\Delta P}{R_f + R_c}. \tag{2}$$

The resistance of the filter cake varies with the amount of material deposited. For a short period of time following a backpulse, the amount of material deposited (F) equals:

$$F = \int_0^t \rho * Q * C dt, \tag{3}$$

where C denotes the concentration of solid in the filtrate. This expression assumes 100% rejection of the suspended solid. The expression also assumes that the solid concentration remains relatively low such that the filtrate flux relatively accurately reflects the volume of slurry delivered to the surface of the filter. Equation 3 then becomes:

$$Q = \frac{\Delta P}{R_f + \alpha \int_0^t \rho * Q * C dt}, \tag{4}$$

where α denotes the specific resistance of a quantity of deposited material. Here α depends on the size and the density of the fouling material. Under conditions of constant pressure drop, reorganization, and differentiation Equation 4 yields

$$\alpha * \rho * Q * C = -\frac{\Delta P}{Q^2}\frac{dQ}{dt}. \tag{5}$$

Integration of Equation 5 produces:

$$\frac{1}{Q^2} = \frac{2\alpha\rho Ct}{\Delta P} + \text{Constant.} \tag{6}$$

Thus, for the case where deposition occurs on the surface of the filter, Equation 6 predicts the behavior during filter cake deposition. However, when filter fouling occurs in the pores of the filter, the filter resistance becomes

$$\frac{1}{R} = \frac{1}{R_f} - \frac{m}{nR_f}, \tag{7}$$

where m denotes the number of pores occluded by deposition and n gives the total number of pores. Under these conditions, the number of pores occluded will vary directly with the amount of material deposited,

$$m = \frac{\beta F}{\rho}. \tag{8}$$

Substitution of Equation 8 into Equation 7 and replacement into Equation 1 produces:

$$Q = \Delta P * \left(\frac{1}{R_f} - \frac{\beta F}{\rho n R_f}\right) = \Delta P * \left(\frac{1}{R_f} - \frac{\beta \int_0^t Q * C dt}{nR_f}\right). \tag{9}$$

Under conditions of constant pressure drop, differentiation of Equation 9 yields

$$\frac{dQ}{dt} = \frac{-\Delta P * Q * C * \beta}{nR_f}. \tag{10}$$

Integration then yields

$$\ln(Q) = \frac{-\Delta P * C * \beta * t}{nR_f} + \text{Constant.} \tag{11}$$

RESULTS

One can analyze filtrate flux data in light of Equations 6 and 11. Figure 2 is a plot of filtrate flux as a function of time for a 1 wt % tetraphenylborate slurry at a constant pressure drop of 20 psi. The lines in Figure 2 provide optimized regressions of Equations 6 and 11 for that data set. Inspection of this figure indicates that Equation 6 (with α = 1330 psi* [gpm/ft^2]/lbm and a constant of 7.22 [gpm/ft^2]$^{-2}$) correlates significantly better with the experimental data (than Equation 11). This result suggests that filter fouling occurs on the surface of the filter during the filtration of tetraphenylborate solids. Note, however, that Figure 2 contains data for only a short period of time following the backpulse. If one extends the time period, a distinct lack of fit between Equation 6 and the experimental data develops. Figure 3 contains a plot of Equation 6, using the value of α and the integration constant as regressed in Figure 2, and experimental data over a 60-min period. This figure indicates that after approximately 7 min of filter operation, Equation 6 increasingly underestimates actual filter performance. Over an extended time, the shearing action of the flow affords a removal mechanism not included in Equation 4 and results in less cake and a higher flux. Therefore, in studying the properties of the filter cake, analysis should be limited to initial data following backpulsing of the filter.

Using only data collected immediately after a backpulse, one can estimate the specific resistance of the deposited filter cake. The authors regressed these values for 30 instances of filter cake development under varying concentration (1 to 10 wt %)and pressure drop (10 to 40psig) conditions using JMP® software version 3.1. Equation 6 does not suggest dependence of α on any of the parameters studied. However, a plot of α as a function of concentration (Figure 4) indicates that α increases as a function of concentration. This result suggests that the specific resistance of the filter cake increases as the solids concentration increases. Note that an increasing solids concentration (through removal of filtrate) was also associated with the length of time the slurry remained in the filtration loop. Therefore, one might attribute this increase to degradation of the slurry particles. The Carman Kozeny equation for flow resistance indicates an inverse square relation for the particle size effect on resistance[3] An increase in the α value from 100 to 800 would correspond to a reduction in particle size by a factor of 2.8. Because of the strong nature of the increase in resistance attributed to the degradation of the particles, this phenomenon warrants further study including particle size determinations for sheared slurries.

The foregoing discussion indicates that one can attribute the fouling of these filters to the deposition of tetraphenylborate solids on the surface of the filter. An additional

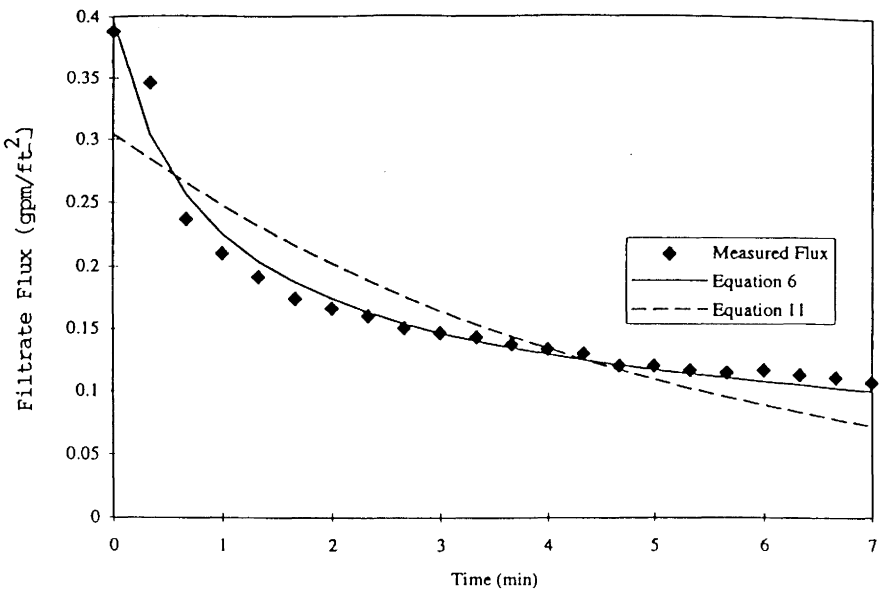

FIGURE 2. Flux vs. time for 1 wt % tetraphenylborate slurry at 20 psi.

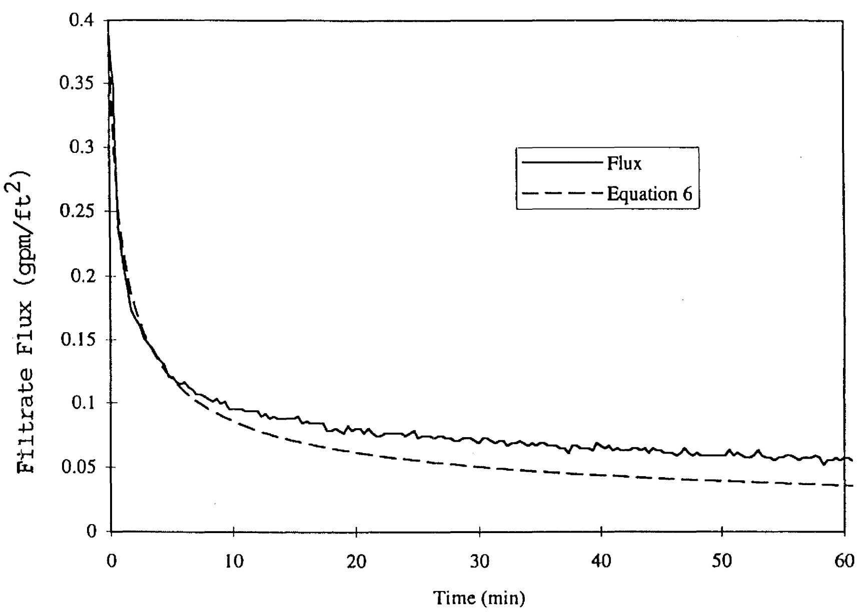

FIGURE 3. Filtrate flux for 1 wt % slurry at 20 psi.

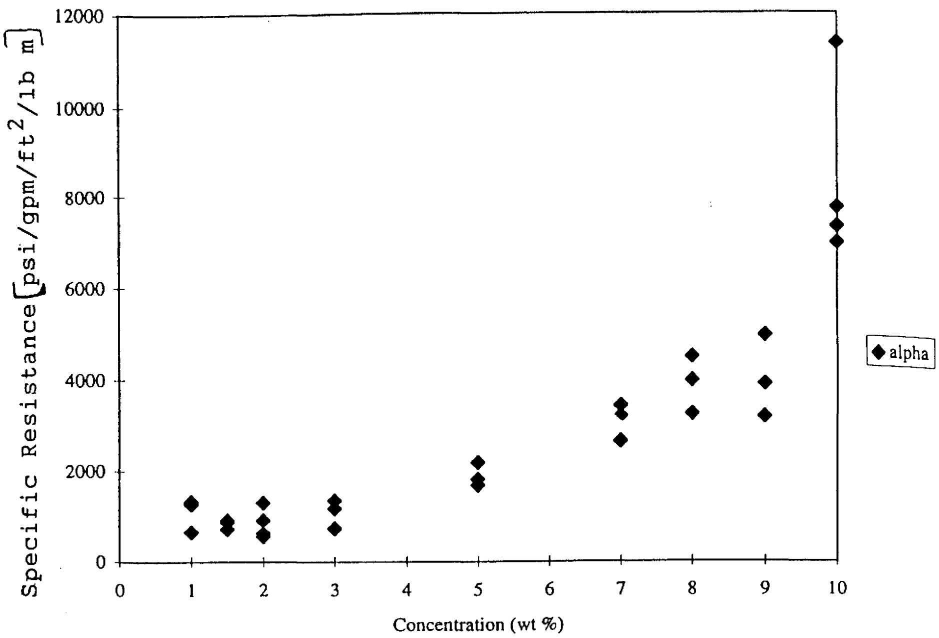

FIGURE 4. Specific Resistance vs Concentration.

question of importance for operation of these filter units involves the removal or reduction of the resistance of the filter cake. As indicated above, following the initial deposition of particles on the filter surface, shear stress at the filter surface begins to mitigate the deposition of further slurry particles. Figure 3 indicates that under these conditions, the filtrate flux remains relatively constant. Since the shear of slurry particles from the surface of the filter inhibits development of the filter cake, increasing the axial velocity of concentrate through the filter might provide an improvement in filter flux.

An additional test established a nearly steady filtrate flux at an axial velocity of 3 ft/s for 3wt % tetraphenylborate slurry at 30 psig. Subsequently, researchers increased the axial velocity to 5 and then 7 ft/s. Figure 5 plots the observed filtrate flux from this test. Though not shown, a slight systematic decrease in the operating pressure occurred at higher velocities. The scatter in the pressure measurements nearly encompasses this

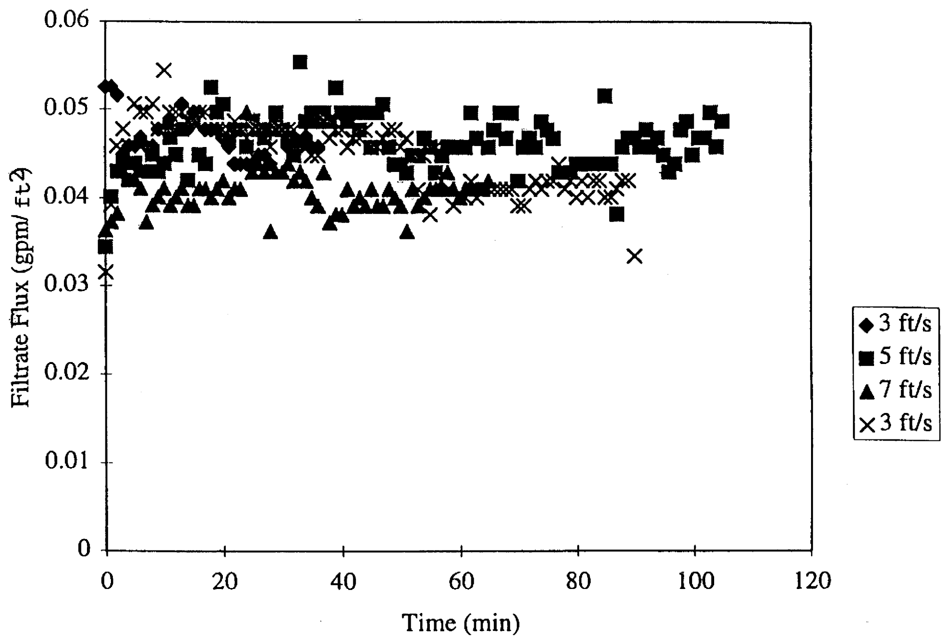

FIGURE 5. Filtrate flux as a function of axial velocity.

trend. Inspection of the figure indicates that the filtrate flux improved temporarily, at most, when the axial velocity was increased. This result suggests that under such conditions, once formed, the filter cake does not erode appreciably due to increasing shear. In fact, inspection of Figure 5 indicates that, if any trend is to be observed, filtrate flux appears to decrease initially when axial velocity is increased. Thus, the ability to rectify fouling by increasing shear is clearly not demonstrated for cakes already in place.

Another test measured the filter flux following backpulses at both 4.5 and 3 ft/s for 3wt % tetraphenylborate slurry at 30 psig. Figure 6 contains a plot of the observed filtrate flux from this test. Inspection of this figure indicates that use of a higher axial velocity during the development of the filter cake results in a higher steady-state filtrate flux. The filtrate flux at 4.5 ft/s proved to be nearly 20% greater than at 3.0 ft/s. Also note that the flux at 4.5 ft/s significantly exceeds any of the fluxes indicated in Figure 5. Table 1 provides the average filtrate fluxes in Figure 5 and the steady-state filtrate fluxes in Figure 6 from 40 to 80 min after the backpulse.

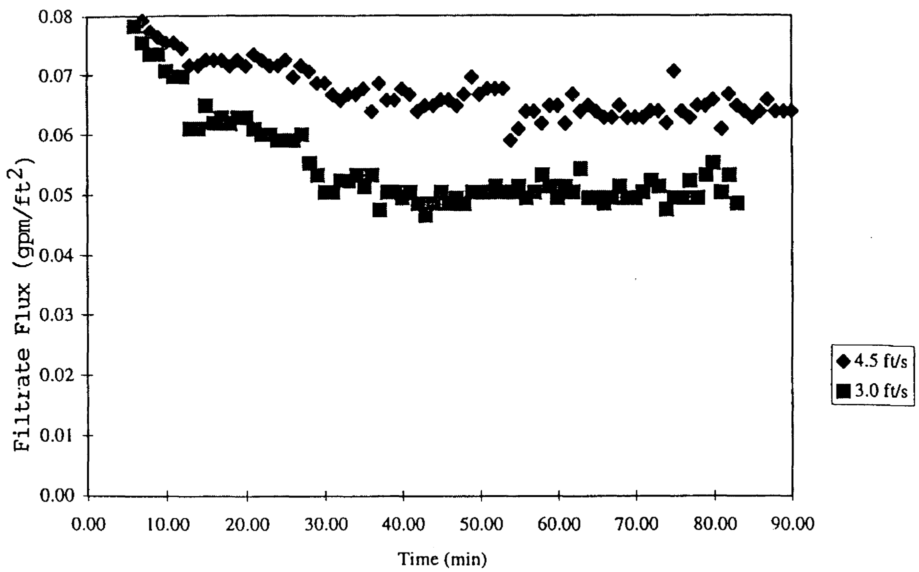

FIGURE 6. Filtrate flux as a function of axial velocity.

TABLE 1. AVERAGE FLUX AND PRESSURE DROP FOR DATA PRESENTED IN FIGURES 5 and 6

Source figure	Velocity (ft/s)	Average flux (gpm/ft^2)	Pressure drop (psi)
Figure 5	3.0	0.047 ± 0.004	31.5 ± 9.5
Figure 5	5.0	0.046 ± 0.006	27.6 ± 7.0
Figure 5	7.0	0.040 ± 0.004	26.0 ± 5.8
Figure 5	3.0	0.045 ± 0.008	30.5 ± 9.6
Figure 6	4.5	0.065 ± 0.004	32.8 ± 0.7
Figure 6	3.0	0.051 ± 0.002	32.1 ± 0.1

Although Figure 5 suggests that increasing the axial velocity will not reduce the resistance of a formed filter cake, Figure 6 implies that the axial velocity under which the filter cake forms can modify the development of fouling by particulates. This dependence likely results from the inhibition of particle deposition by the increased shear. The influence of shear would prove more effective at lower fluxes.

The observation that an increase in axial velocity does not change the resistance of a formed filter cake is neither universal nor intuitively obvious. However, sodium and potassium tetraphenylborate slurries exhibit a large yield stress, which increases as the solids content in the slurry increases, reaching approximately 25 N/m^2 for 10 wt % slurries.[4] Thus, it is likely that once formed, the filter cake will be held intact by this yield strength.

ITP Performance

During preliminary facility testing, the ITP filtration process operated for a short period . During those operations, facility personnel concentrated the slurry from approximately 1 wt % to 3 wt % by removal of filtrate. Filtrate production was set to either 50 or 20 gpm, based on production for the entire filtration unit. (This production rate corresponds to a filtrate flux of approximately 0.1 to 0.25 gpm/ft^2.) As filter resistance increased, personnel raised the operating pressure to maintain the filtrate production rate. Figure 7 contains a plot of the operating pressure as a function of time for 2 wt % slurry. Prior to the period of time indicated, the filter surface was washed of cake through the backwash of filtrate. The filter cake accumulated at a filtrate flow rate of 50 gpm for 3 h. Note that this period greatly exceeds that required to establish the filter cake in laboratory tests (5 min). Previous experience with ITP fluid demonstrated a considerable difference with the unirradiated laboratory test fluid.[5] After 3 h, the filtrate flow rate and operating pressure were decreased in three steps over a period of approximately 1 h. Under the lower pressure conditions, the filtrate flow rate and operating pressure proved stable for at least 2 h. This result suggests that the axial velocity prevented the further deposition of TPB solids at the lower filtrate production rate, while the shear on the filter cake failed to remove cake from the filter surface.

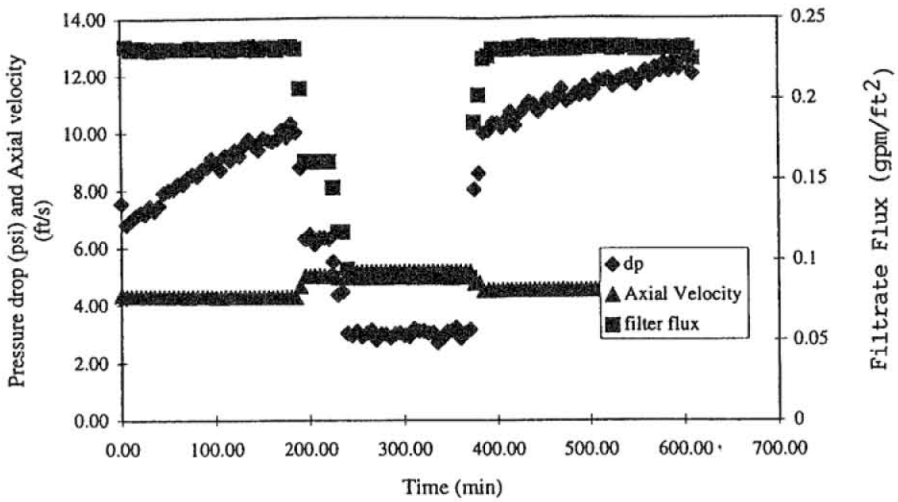

FIGURE 7. ITP filter performance.

Subsequently, personnel increased the filtrate production rate to 50gpm. The required operating pressure effectively equaled that originally required prior to reducing the filtrate flow rate. This result also suggests that the filter cake remained unchanged by operating at the lower pressure and slightly higher shear rate.

CONCLUSIONS

Laboratory tests investigated cross-flow microfiltration of slurries of 1 to10 wt % sodium tetraphenylborate under conditions approximating ITP processing. The results show, for filter operations interspersed with backpulse cleaning regimen, a reduction in flux consistent with the model of layer formation and inconsistent with pore occlusion or in-depth plugging. Further, as the flux continues to decline, the excluded solids fail to contribute to further layer formation and a steady operating flux results. A material balance insists that the excluded solids continuously escape from the filter surface.

Tests of filter cakes established at low shear and sequentially subjected to higher shear rates showed a slight, but temporary, change. The shear did not remove the cake to allow a higher, steady flux. In contrast, filter cakes established at different shear conditions display systematically different steady asymptotes. In combination with the previous observation, this suggests that shear inhibits, but does not remove, the layers of filter cake. This observation appears reasonable, considering the tetraphenylborate slurry exhibits Bingham plastic behavior with a yield stress higher than the applied fluid shear stress.

Brief testing of the ITP filter system indicated, as observed previously, that the fouling resistance was less severe than that exhibited by the laboratory test fluid. Similar to the laboratory test fluid, though, there was an increase in resistance indicative of a filter cake buildup. When reduced flux, caused by lowered operating pressure, was imposed on the established cake, a period of steady operation was observed. This steady operation indicated that neither an increase in cake nor a removal of cake occurred; thus, a balance of shear removal and filter concentration was achieved.

LIST OF SYMBOLS

α	specific resistance	psi/(gpm/ft^2)/lb m
β	pore plugging constant	1/ft^3
ρ	filter cake density	lb m/gallon
C	slurry concentration	lb m/lb m
F	solids deposited	lb m
m	number of pores plugged	
n	total number of pores	
ΔP	pressure drop	psi
Q	flux	gpm/ft^2
R	resistance	psi/(gpm/ft^2)
t	time	min

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