Membrane Plugging Mechanisms Drive Smart Filter Selection Strategies

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INTRODUCTION

Liquid filtration techniques are used across diverse industries to increase overall equipment efficiency (OEE) through the removal of various contaminants: particles, metals, organics, and gels. As defect analysis techniques improve and identify even smaller contaminants, killer contaminant size also reduces. Highly retentive filters are therefore required to capture smaller particles – a task that is not always straightforward.

Traditionally, increasing a filter's retention rating will both increase pressure drop and reduce filter lifetimes, resulting in increased process downtime. Extending filter lifetime requires a careful balance between particle retention and pressure drop. Therefore, it is essential to understand a fluid's contaminant concentration as it enters a filter.

Grades of chemistry can range across industries. Typically, electronics grade chemicals have low particle levels when packaged by the manufacturer, but there are many opportunities along the supply chain where clean chemistries can become contaminated. These opportunities include:

- Transporting the chemical to the site
- Connecting from the bottle, drum or tank car to the system piping
- Moving the fluid through piping, valves, pumps and regulators
- Processing in recirculating flows
- Interrupting the system

When the process chemistry becomes particularly saturated with contaminants, the filter can plug, increasing pressure drop (in constant flow rate systems), decreasing the flow rate (in constant pressure systems), or a combination of the two.

The purpose of this Applications Note is to provide a basic understanding of plugging mechanisms to ultimately provide guidance when selecting filters and designing optimized filtration schemes.

MEMBRANE PLUGGING MODELS

Three models are defined to understand plugging mechanisms: Cake formation, complete pore plugging, and gradual pore plugging.

Cake Formation Model

Cake formation normally occurs when particles larger than the surface pore size accumulate on the membrane surface, forming a "cake" (see Figure 1). This cake layer provides an additional porous surface through which the liquid must pass. As a result, the cake may increase the particle removal efficiency of the filter, while also increasing a filter's resistance.

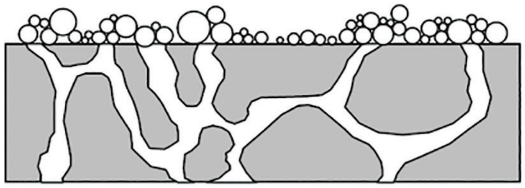

Figure 1. Pictorial view of cake formation

Figure 2 shows typical performance curves that characterize cake formation. Curves are shown for both constant pressure and constant flow rate processes. For constant flow, the increase in pressure drop is linear with time. For constant pressure, most of the flow decreases early in the process.

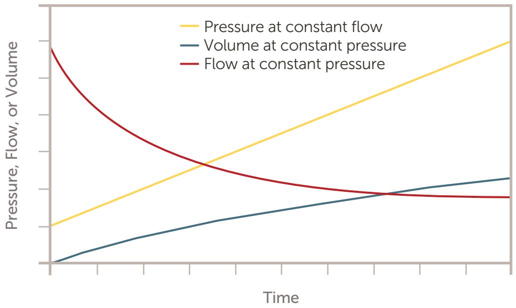

Figure 2. Example of typical cake formation system curves

Complete Pore Plugging Model

Figure 3 depicts complete pore plugging. This typically occurs when the particle sizes are similar to the mean pore size. In this model, particles block individual pores. As individual pores are plugged, flow is diverted to other pores that successively plug. Eventually, this reduces the available membrane area and increases the membrane's resistance.

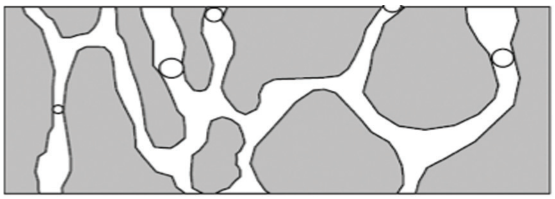

Figure 3. Pictorial view of complete pore plugging

Figure 4 provides examples of typical complete pore plugging curves. Curves are shown for both constant pressure and constant flow rate processes. Unlike in cake formation, the curves are not linear. At the end of the filter life, pressure drop increases dramatically.

For complete pore plugging, the number of open pores is reduced as pores plug. As a result, either the flow rate is reduced, or the pressure drop increases.

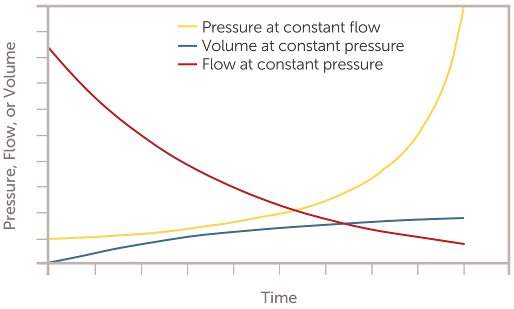

Figure 4. Typical complete pore plugging system curves

Gradual Pore Plugging Model

Gradual pore plugging is most dominant when retained particles are smaller than the pores. In this case, particles in the fluid approach the membrane, enter the pores, and adhere to the pore walls through one of many adsorption mechanisms, as illustrated in Figure 5. Unlike the complete pore plugging model, pores do not become completely blocked. In this case, the adhesion of contaminants to the walls decreases the available pore diameter and increases the resistance of the membrane.

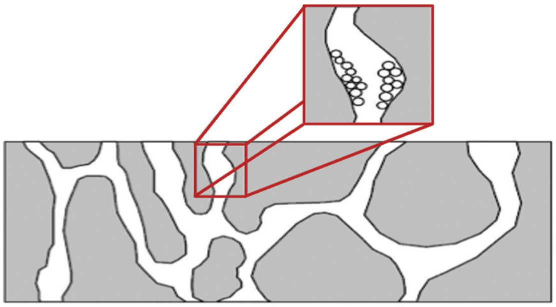

Figure 5. Pictorial view of gradual pore plugging

From typical test data shown in Figure 6, it is very difficult to determine if a filter is plugging due to complete or gradual pore plugging as these curves are also non-linear. Like complete plugging, pressure drop increases dramatically at the end of filter life.

As pores plug in gradual pore plugging model, the number of pores available for flow remains constant while the pore diameter decreases. As a result, small changes in the pore diameter can result in large changes to either the flow rate or the pressure drop.

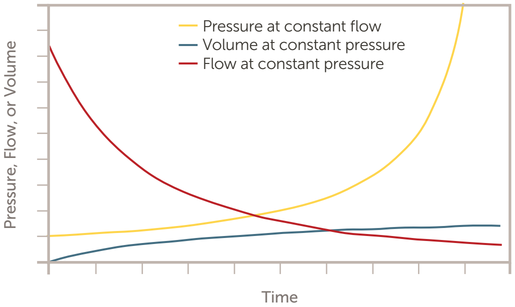

Figure 6. Typical gradual pore plugging system curves

All the mechanisms defined are summarized in Table 1.

Table 1. Filter plugging mechanisms summary

	Cake formation	**Gradual pore plugging**	**Complete pore plugging**
Typical particle size	Larger than the surface pore size	Smaller than the mean pore size	Similar to mean pore size
Mechanism effect on total filtration surface area	Increases	Neutral	Decreases
Mechanism effect on flow rate	High reduction	Slow reduction	Moderate reduction
Mechanism effect on fluid flow path	Fluid diverted away from blockage	Fluid diverted to increasingly smaller channels	Fluid diverted away from blockage

Identifying the Plugging Mechanism

In order to best identify the method of filter plugging in a system, characteristic equations (Table 2) can be applied to empirical data to identify the best fit for the data.

Table 2. Equations defining plugging mechanisms*

Model	**Characteristic equation**	**Constant pressure**	**Constant flow**
Cake formation	$\frac{dV}{dt} = \frac{\Delta PA}{\mu\left(\frac{\alpha\beta V}{A} + R_m\right)}$	$\frac{t}{V} = C_1V + C_2$	$\Delta P = C_3t + C_4$
Complete pore plugging	$\frac{dV}{dt} = \frac{\pi\Delta PA}{8\ \mu L} r_p^4 N'_{p0} - \frac{\pi\Delta PA}{8\ \mu L} r_p^4 k_2 V$	$\frac{dV}{dt} = C_5 - C_6V$	$\frac{1}{\Delta P} = C_7 - C_8t$
Gradual pore plugging	$\frac{dV}{dt} = \frac{\Delta PA}{8\ \mu L} r_p^2 N'_p$	$\frac{t}{V} = C_9t + C_{10}$	$\frac{1}{\sqrt{\Delta P}} = C_{11} - C_{12}t$

**Derivation of all equations can be found in the original manuscript, "Understanding Membrane Plugging Mechanisms," Mykrolis applications note, MAL116 (2000).*

- A Membrane surface area (m^2)
- C_x Constant
- k_2 Number of plugging particles per volume of filtrate (m^{-3})
- L Membrane thickness (m)
- M_0 Initial system flow rate (m^3/s)
- Np Number of pores still open
- N_{p0} Number of pores in a clean filter
- N'_{p0} Number of pores/unit area in a clean filter (#/m^2)
- ΔP Pressure drop across the membrane (N/m^2)
- $\Delta P0$ Initial pressure drop across the membrane (N/m^2)
- R_m Membrane resistance (m^{-1})
- r_p Average pore radius (m)
- t Time (s)
- V Volume of filtrate that passes through the filter (m^3)
- μ Fluid viscosity (Ns/m^2)
- α Specific resistance of the cake that forms on the membrane surface (m/kg)
- β Mass of particles per volume of filtrate (kg/m^3)
- ρs Mass density of the plugging particles (kg/m^3)

FOR CONSTANT PRESSURE DROP SYSTEMS:

- Both the cake formation and gradual pore plugging characteristic equations are linear. Plotting t/V versus both volume and time will determine which of the two models more accurately reflects the actual process. Performing a linear regression on the two curves can provide more confidence, although it may be necessary to delete the first few points if it can be determined that measurement error is involved.
- The complete pore plugging characteristic equation is also linear. To determine if this model more accurately reflects the process, plot flow rate (dV/dt) versus volume. If complete pore plugging is the dominant method, flow rate versus volume should be linear with a decreasing slope.

FOR CONSTANT FLOW RATE SYSTEMS:

- All the characteristic equations are linear. By plotting ΔP, ΔP^{-1}, or $\Delta P^{-1/2}$ versus time, it can be determined which of the models more accurately reflects the actual process. Again, a linear regression can provide more confidence.

Comparison of Models to Experimental Results

Table II presents data taken during testing of an ultra-filtration membrane (molecular weight cut-off approximately 200,000 Daltons). The test was conducted using deionized water (DIW), applied at a constant pressure flowing through a 47 mm disc.

Figure 7 provides a graphical view of the data from Table 3. This data will be examined using the characteristic equations from Table 2 to determine the method of plugging.

Table 3. Experimental data set #1

Time	Volume
0.25 hr	2 L
0.5 hr	5 L
1 hr	7 L
2 hr	13 L
4 hr	20 L
12 hr	40 L
24 hr	60 L
32 hr	70 L
48 hr	88 L
72 hr	110 L

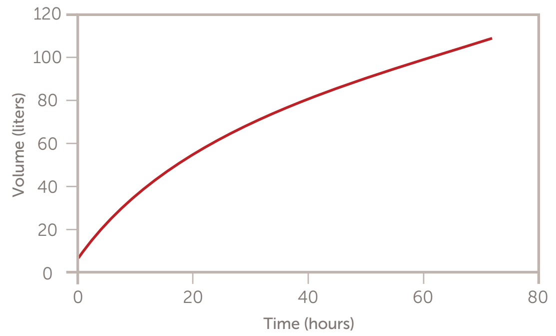

Figure 7. Graphical representation of data set #1

Figure 8 shows the data from Table III plotted as t/V versus V to determine if the data agreed with the cake formation model. A linear regression was performed. The high correlation coefficient (0.997) indicates that the filter is most likely plugging via cake formation.

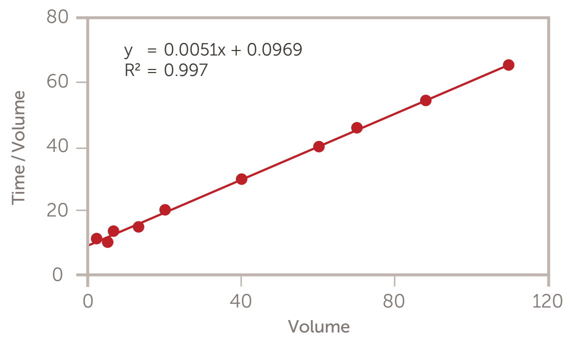

Figure 8. Match to cake formation

Figure 9 shows the data from Table 3 re-plotted as t/V versus t to determine if the data agreed with the gradual pore plugging model. A linear regression was performed, resulting in a lower correlation coefficient (0.9475) than Figure 8. Also, the arcing of the data points indicates that although some gradual pore plugging may be occurring, cake formation is much more dominant.

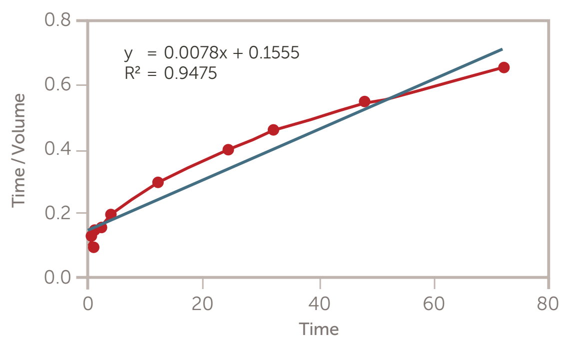

Figure 9. Match to gradual pore plugging

Figure 10 shows the data from Table 3 re-plotted as flow rate versus filtrate volume to determine if the data agreed with the complete pore plugging model. The lower correlation coefficient (0.5888) and non-linearity of the regression indicates that complete pore plugging does not appear to be a factor.

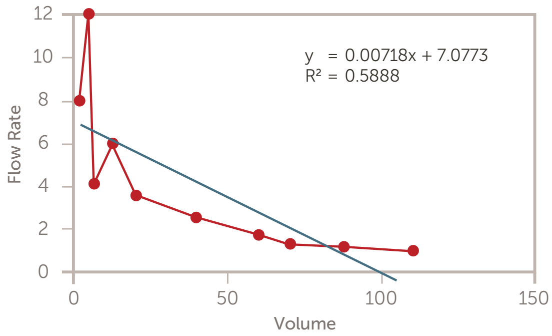

Figure 10. Match to complete pore plugging

This example showed how one might use the models to predict the method of plugging in this system. For an ultrafiltration membrane with an extremely small pore size, we would expect that it would plug via the cake formation model by accumulating particulates on the upstream side of the membrane.

Identifying Smart Filtration Strategies Based on Plugging Mechanism Studies

We can identify the type of plugging that occurs with a specific membrane-fluid combination. Using that information, how can we provide improved filtration and/or longer lifetime?

Increasing membrane area or adding prefilters are common ways to improve throughput, but knowing the type of plugging helps predict the magnitude of the effect of added area and which type of prefilter to use.

INCREASING FILTER AREA

Increasing filtration membrane area in a system that experiences filter caking results in two dramatic effects: A thinner cake layer across the surface area and reduced fluid velocity through the membrane and cake.

If the resistance of the cake layer is large compared to the resistance of the membrane, throughput is a function of the square of the area. Doubling the area results in a throughput increase of four times. If the allowable pressure drop increase is modest, for example by a factor of two from the initial pressure drop, the final cake resistance will be equal to the membrane resistance. In this situation, doubling the area will result in a six-fold increase in throughput.

Therefore, increasing area for a cake plugging system is a good strategy under any circumstance.

An additional benefit of increased area in systems that experience filter caking is that the face velocity decreases. A lower velocity gives small particles a more time to travel through the filter without getting trapped in the forming cake. If the concentration of particles is reduced by half, it is likely that more than twice the volume could be filtered.

For both complete and gradual plugging at the limit of a fully plugged filter, increasing area has a linear effect on throughput. Since doubling area doubles the number of available pores, the throughput only increases by a factor of two when membranes are fully plugged. If the system is not allowed to fully plug, increasing the area can have more than a linear effect for complete and gradual modes.

For example, imagine a completely plugged system where the initial pressure drop is 1 psid and the allowable final pressure drop is 2 psid. Therefore, half of the pores plug. Doubling the area in this condition will triple throughput. All the pores in the extra area will plug before reaching the final pressure drop of 2 psid.

ADDING PREFILTRATION

Prefilters are less retentive filters placed before the final filter. They are designed to extend the life of final filter by reducing particle load. The mechanism of final filter plugging informs the desired attributes of the prefilter.

If the final filter plugs by a gradual mechanism, the particles are small and are being removed by an adsorptive mechanism. The surface of the prefilter should be of a similar nature to the final filter, but the pores should be larger. The prefilter should be relatively thick so that it has a chance to remove the particles by increasing the residence time. Since the pores are larger, this will not have a significant effect on the pressure drop.

In cake and gradual plugging, a more open prefilter will improve final filter performance. Prefiltration will remove the largest particles, reducing the mass of particles per volume of filtrate. Prefilters will reduce the mass of particles since the larger particles still have significant mass. It has been shown that filtrate particle size distributions are linear on a log-log plot of particle concentration vs. particle diameter,[1] and this slope of this line is typically between -2 and -3. Typical example fluids are shown in Figure 11. Although either filter lifetime will be extended, or throughput volume will increase, the improvement is neither linear nor is it as effective as adding additional filtration area.

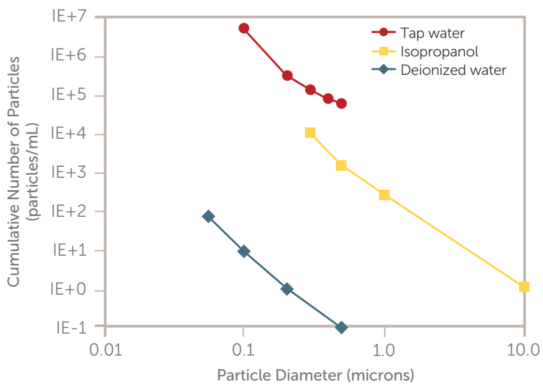

Figure 11. Measured size distribution of particles in liquids[2]

For cake plugging, it is also important to note that although prefilters will reduce the mass of particles per volume of filtrate, they will also increase the resistance of the cake layer. The Carman-Kozeny equation tells us that the cake layer resistance is proportional to the inverse of the square of the average particle size in the cake.[2] As larger particles are removed, the average particle size decreases, and as a result, the resistance of the cake layer increases exponentially. However, as the larger particles are removed, the mass of particles per volume of filtrate decreases more rapidly than cake resistance increases. As an example, reducing the mass of particles per volume of filtrate by 80% causes the cake resistance to increase by 36%, resulting in a net product reduction of 73%.[3]

For complete pore plugging, adding a less retentive prefilter that removes particles by adsorption is the only practical solution. Since the particles are the same size as the pores of the final filter, a prefilter that operates with complete pore plugging would be a duplicate of the final filter. Therefore, a more open prefilter operating with gradual pore plugging is the remaining choice. Choosing the surface of the prefilter so particles are removed by adsorptive mechanisms is the best choice when the final filter plugs by the complete mechanism.

Conclusions

Understanding the retention mechanism of filtration in your system is an important first step in choosing filtration solutions that can extend filter lifetime and increase your operational productivity. Three models for membrane plugging, namely cake formation, complete pore plugging, and gradual pore plugging, have been identified. Using characteristic equations, you can analyze empirical data to confirm the method of plugging. There are many variables available to improve throughput and increase filter lifetime, some of which can be controlled by changing the fluid, changing the system variables, and making different filtration choices. With an understanding of the plugging method, filtration recommendations, such as those in Table 4, can be made to improve throughput and increase filter lifetime.

Table 4. Recommendation to improve filter throughput

Plugging model	Constant pressure process	Constant volume process
Cake filtration	Increase membrane area	Increase membrane area
Gradual pore	Increase membrane area Increase membrane thickness	Increase membrane area Increase membrane thickness and area
Complete pore	Increase membrane area Decrease pore size	Increase membrane area Decrease membrane thickness

References

[1] Lee, J., et al., *Filtration of Real-World Particles in Liquids by Microporous Membrane Filters*, Journal of the IES, May/June 1995, p. 20.

[2] Ho, W.S. and Sirkar, K., *Membrane Handbook*, 1992. p. 463.

[3] Lee, Jae-Keun, Liu, Benjamin Y.H., Rubow, Kenneth L., and Yoo, Seong-Ho, *Filtration of Real-World Particles in Liquids by Microporous Membrane Filters*, Journal of the IES, May/June 1995, p. 20.

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