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BIOFOULING OF MICROFILTERS AT THE SAVANNAH RIVER SITE F/H-AREA EFFLUENT TREATMENT FACILITY

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ABSTRACT

The F/H-Effluent Treatment Facility uses state-of-the-art water treatment processes to remove contaminants from low-level radioactive wastewater at the Savannah River Site. The plant replaces seepage basins that were closed to comply with the 1984 amendments to the Resource Conservation and Recovery Act (RCRA). The facility removes both radioactive and nonradioactive contaminants from the effluents originating from onsite waste management facilities. The unit processes involve filtration, ion exchange, activated carbon absorption, and reverse osmosis. The filtration step is prone to considerable fouling, reducing the overall throughput of the facility.

The filters utilized in the process are Norton Ceraflo™ ceramic microfilters. It was discovered that bacteria were primarily responsible for the severe filter fouling. Inorganic fouling was also observed, but was not normally as severe as the bacterial fouling. The bacteria densities necessary to induce severe fouling were not significantly higher than those often found in surface water streams. Diversion of waste streams containing the highest quantity of bacteria, and various methods of source reduction were implemented, which dramatically improved the filter performance. Addition of aluminum nitrate at low pH further improved the filter performance.

INTRODUCTION

The Savannah River Site (SRS) constructed an effluent treatment facility (ETF) to treat wastewater containing low levels of radionuclides and hazardous chemicals. The facility operates under a National Pollution Discharge Elimination System (NPDES) permit by the South Carolina Department of Health and Environmental Control (SCDHEC) to discharge treated water. The wastewater originates from various processes, primarily: (a) waste evaporator overheads, (b) contaminated cooling water, and (c) storm water runoff. The evaporator overheads are the predominant source of influent, with the other sources being nonroutine. The ETF began operation in October of 1988 and replaced seepage basins for disposal of these waste streams. Through January 1, 1992, the facility has processed over 67 million gallons of wastewater, and has never exceeded any limits on the NPDES permit.

PROCESS DESCRIPTION

Routine influent to the ETF contains small quantities of many soluble and sparingly soluble salts. The predominant component in the wastewater is normally sodium nitrate, from the neutralization of nitric acid with sodium hydroxide. Small quantities of iron (III), aluminum (III), and silica comprise the bulk of the insoluble solids. Various metal ions, both radioactive and nonradioactive, are present (Table I), as well as low levels of organic compounds (primarily tributyl phosphate from separations processes). The quantities of these species varies considerably in day-to-day operations.

(Place Table I here)

The goal of the ETF is to reduce the volume of the waste that must be disposed of by concentrating the hazardous components. The majority of the concentrated waste is disposed of in saltstone, a composite of concrete and salt-containing waste that is contained in concrete vaults. Small quantities of other waste are also generated in the ETF, including ion exchange resin and granular activated carbon that are disposed of as low-level nonhazardous waste. Treated water is discharged through the NPDES outfall to a surface water stream. The discharge criteria include limits on oil and grease, biological oxygen demand, and many toxic metals. Release guidelines also dictate that the radioactive metal ions be removed to a very low level (<4.5 pCi/mL gross beta/gamma, <1 pCi/mL gross alpha).

The individual process steps were designed to remove the regulated components to well below the discharge limits. The individual treatment steps (Figure 1) are:

- pH adjustment
- microfiltration (MF)
- mercury-specific ion exchange
- activated carbon for organic removal
- reverse osmosis (RO)
- polishing ion exchange (with regeneration)
- evaporation

(Place Figure 1 here)

The flowsheet of the process is shown in Figure 1. Neutralization of the pH is conducted in a two-step process with dilute (5 wt%) sodium hydroxide and nitric acid. Metal hydroxides and other suspended solids are precipitated in the pH adjustment step and are removed by filtration in order to eliminate settling downstream and to minimize the solids loading on the RO. The microfiltration step utilizes crossflow Norton Ceraflo™ ceramic filters to concentrate the solids to 1% of the initial volume. A mercury-specific ion exchange resin (Duolite GT-73) is used to extract mercury ions and prevent mercury absorption on the carbon beds. Activated carbon beds are utilized to absorb organic compounds and prevent organic fouling of the RO. The reverse osmosis unit (Filmtec high-rejection spiral-wound membranes) concentrates the soluble salts, both hazardous and nonhazardous. The RO operates at 90% recovery to minimize the load on the evaporators. A polishing ion exchange resin (Mitsubishi Diaion HPK-25) removes the remaining radioactive metal ions. Clean water is collected in hold tanks and tested for compliance prior to discharge. The concentrate streams from the RO and MF, and the ion exchange regenerate solution are further volume-reduced in the evaporator. The evaporator overheads are returned to the process for treatment prior to discharge. Concentrated waste from the evaporator bottoms is pumped to a hold tank prior to disposal in the saltstone process.

The influent wastewater can vary markedly in its composition. As a result of this variability, each of the process steps must be capable of removing contaminants while maintaining a reasonable throughput. The microfiltration step is particularly susceptible to changes in influent conditions. Temperature, solids concentration, pH, and organic content affect the filter performance. The other process steps are more robust and are more capable of handling the variability in the waste stream.

MICROFILTER DESCRIPTION

The filters are Norton Ceraflo™ microfilters that are constructed of sintered alpha alumina. The nominal pore size is 0.2 microns. A single multilumen filter element is approximately 80 cm long, 2 cm in diameter, and has 19 flow channels. Ten multilumen elements are bundled together with a steel connector, and two bundles are in each housing. Each skid contains four housings, connected in parallel. Three skids in series comprise one filter unit, for a total of 240 filter elements with a total filter surface area of approximately 37 m^2. The filters are operated in crossflow mode with a linear velocity of 2.5 m/sec, and an average transmembrane pressure of 30 psi. The filters can be backpulsed with filter permeate at a transmembrane pressure of 30 psi.

A laboratory-scale demonstration unit was constructed to enable research on fouling by various constituents. This unit contains three single-channel filter tubes connected in series. Transmembrane pressures and crossflow velocities are similar to those in the full-scale unit.

MICROFILTER FOULING

The original design basis for the filters required a 100 gpm average flow rate through each filter. Upon startup of the facility, actual flow rates were significantly less than 100 gpm, and the run time between chemical cleanings was very short (Figure 2). In addition, much of the waste required direct evaporation due to its severe fouling potential. This filter fouling severely impacted the ability of the ETF to process the wastewater from the separations areas. After several months online, further degradation in the filter performance was observed. This event correlated with shutdown of several upstream facilities that reduced the influent flow rate and changed the typical stream composition.

(Place Figure 2 here)

Initial research into the fouling problems focused on the inorganic constituents in the wastewater. Laboratory-scale demonstration tests showed that constituents such as colloidal silica could cause severe filter fouling. Examination of the actual influent wastewater did not reveal that silica or other inorganic constituents were responsible for the fouling. Initial sampling revealed very low levels of bacteria. Filter fouling and inorganic composition did not correlate, and the filter performance was unrelated to total suspended solids content.

Further investigation into the biological activity in the influent revealed that significant densities of bacteria were present in the influent wastewater. The quantity of bacteria that would cause filter fouling was unknown. A study was initiated to quantify the impact of bacteria on the filter performance.

Laboratory investigations were initiated to determine if prefiltering the influent would increase filter performance, or if an additive could be found that would minimize the filter fouling. It was determined that the best method for remediating the bacterial fouling was to add aluminum nitrate (10 to 20 mg/L) at low pH (<2.7) to the bacteria-containing water.

EXPERIMENTAL

Influent wastewater and process samples are collected as grab samples in sterile bottles. The samples are analyzed by the acridine orange direct count[1] (AODC) technique, a 10-microliter aliquot of the sample is placed on a microscope slide and allowed to dry. The fixed samples are then stained with a fluorescent dye (acridine orange). Since this stain attaches only to nucleic acids found in biological material it greatly aids discrimination of bacteria amongst other debris. Fluorescins bacteria are then counted directly using an epifluorescence microscope.

All bacteria density averages are reported as averages of the log of the densities. The error reported is the standard error of the mean. The minimum detection limit for the method is 9.00 x 10^4 bacteria/mL.

Simulant solutions were prepared from deionized water for the laboratory-scale unit. Inorganic salts, iron nitrate, aluminum nitrate, and sodium metasilicate were used as the predominant inorganic constituents present in the wastewater. Bacteria isolated from the ETF were cultured in a nutrient broth, centrifuged, and washed prior to addition to the simulant solution.

RESULTS

Bacteria density in influent samples have been collected routinely since 10/3/89. Their effect on performance of the Norton filters is shown in Figure 3. The filter performance is categorized as poor (<40 gpm average), medium (40 to 75 gpm), and good (>75 gpm). The average of each of the three categories was determined, and the standard error of the mean calculated. The standard error of the mean is indicated on Figure 3.

(Place Figure 3 here)

Various influent point sources were examined for bacteria densities. Significant bacterial growth was observed in wastewater tanks that were near neutral pH and were retained several days before discharge. It was also observed that tank heels could be significant sources of bacteria, presumably because of settling.

Bacteria densities were also determined at several points within the ETF system. Most bacteria (0.2 to 3 microns) will not pass through the filter since the filters are rated to have a nominal pore size of 0.2 microns. This was confirmed by sampling immediately downstream of the filters. Only small quantities of bacteria were identified in the filter permeate, which were attributed to growth within the system after filtration. Effluents from both the Duolite GT-73 Hg-removal resin columns and activated carbon columns, which are immediately downstream of the filters, were found to contain high densities of bacteria. It was established that significant biological growth was occurring in the resin columns and carbon columns.

The addition of aluminum nitrate increased the filterability of some waste streams dramatically (Figure 4). Waste streams that contained bacteria and could not be diverted to an evaporator could be filtered after the addition of small amounts of aluminum nitrate (10 to 20 mg/L Al(+3)) at low pH (<2.7). This addition has little impact on the other unit processes or discharge water quality.

(Place Figure 4 here)

Laboratory-scale tests have demonstrated that lysed bacteria will foul the filters virtually as rapidly as intact bacteria (Figure 5). Cells that are lysed with sodium hydroxide solution are undetectable by the AODC bacteria cell counting technique, but cause rapid filter flux loss.

(Place Figure 5 here)

DISCUSSION

It is clear from Figure 2 that bacteria play a key role in filter fouling. At high bacteria densities (>2.0 x 10^6 bacteria/mL) filter flow rates are always less than 40 gpm. Filter performance never exceeds 75 gpm if intermediate densities of bacteria (2 x 10^5 to 2 x 10^6 bacteria/mL) are present. Occasionally, low densities of bacteria (<2 x 10^5 bacteria/mL) also yield poor filter performance. This is normally attributable to inorganic fouling, inadequate cleaning cycles, or lysed bacteria.

The most effective method of remediating the effect of bacteria was to eliminate the point sources that contained the highest densities. This was done in May 1990, and was concurrent with a sanitization of the process. The point source that contained the most bacteria was a resin flush solution from the Duolite GT-73 columns that had been routed back to the influent feed tank. This stream was diverted directly to the evaporators to eliminate its effect on the filter. Other upstream point sources, such as tank heels, were also eliminated or reduced. Elimination of the point sources was very effective, and was primarily responsible for the sudden increase in filter performance in May 1990 (Figure 2). Although it is not indicated in Figure 2, the filter run duration also increased dramatically.

Although point source elimination improved filter performance, it is not possible to eliminate all of the bacteria. The addition of aluminum nitrate has been successfully tested in the ETF to mitigate the bacterial fouling. These tests produced significantly higher flow rates, enabled the cleaning frequency to be lowered, and improved the effectiveness of the filter cleaning cycles (i.e. the startup flow rates after cleaning were higher). The addition of aluminum nitrate to the influent wastewater was tested on the ETF filters (Figure 4). This wastewater was determined to contain intermediate densities of bacteria (8.0 x 10^5 bacteria/mL). During these tests, it was also observed that sudden, severe filter fouling would occur if the pH of the filter influent was not maintained at 7.5 ± 1.5 pH units.

It is not possible to simply use a biocide in the influent to prevent bacterial growth for several reasons. The origin of the bacteria, in most cases, was at the point sources and only minimal growth occurred once the water was received in the waste collection tanks. It has been shown that dead bacteria foul the filters as quickly as live bacteria, so biocide addition after growth was ineffective. The typical oxidizing biocides such as chlorine are also reactive towards the Duolite GT-73 resin, rendering the resin less active in mercury removal. Due to high organic content and high ammonia content, the chlorine demand of the wastewater could introduce sufficient chlorine to exceed the chloride limit on the waste concentrate. However, it was found that the Duolite GT-73 could be effectively cleaned with dilute sodium hydroxide solution, minimizing the bacterial growth on the resin.

Fouling due to inorganic components has also been observed. This does not appear to be as severe as fouling from biological material but can be significant, particularly when high concentrations of aluminum are present. Upsets in the pH of the wastewater can have severe consequences on filter performance. It has been shown that high quantities of silica can foul the filters rapidly.

The technique utilized for bacteria enumeration (AODC) identifies both dead and live bacterial cells, and does not distinguish them from one another. Most of the bacteria present in the influent and within the ETF were Gram-negative rods, which are common in wastewater treatment plants. The exterior of these bacteria are coated with an adhesive layer composed primarily of lipopolysaccharide (LPS). The LPS is known to be involved in bacterial biofilm formation, and may be involved in preconditioning film formation.[2] The LPS, and other biomolecules, are released when cells are lysed (disintegrated) and are not easily degraded. The AODC analysis stains nucleic acids and therefore does not identify or quantify other biomolecules. The quantity of biomolecules present in the ETF wastewater is not currently known but may play a significant role in filter fouling.

CONCLUSIONS

The impact of bacteria on filter performance is quite severe; bacteria densities over 2.0×10^6 bacteria/mL normally result in filter flow rates of less than 40 gpm. Diversion of the point sources that contained the highest densities of bacteria significantly reduced the influent bacteria densities and increased the filter flow rates. Further improvement in filter performance can be obtained by the addition of aluminum nitrate to the influent wastewater. Maintaining the pH at a constant value is imperative to the success of aluminum nitrate addition. It is now estimated that the facility will be able to meet the demand for wastewater treatment from routine influent sources for the forseeable future.

ACKNOWLEDGMENT

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2. *Advances in Microbial Ecology*, 6, K.C. Marshall, Ed, Plenum Press, pp 199-230.

Tabl Averageuentuent Concentrations

Comnt	nc (mc (mg/L)	Component	Conc	(unit)
ɿ	⊦ ⊦	Ba	0.04	(mg/L)
(	⌉ ⌉	Cr	0.04	(mg/L)
(	4 4	Fe	2.6	(mg/L)
]	)63)63	K	0.8	(mg/L)
]	¦2 ¦2	Mn	0.18	(mg/L)
]	))	Ni	0.092	(mg/L)
]	. .	U	1.8	(mg/L)
;	⌉ ⌉	Cl^-	1.2	(mg/L)
(	))	F^-	1.1	(mg/L)
NH_4^+		NO_2^-	1.6	(mg/L)
]	))	PO_4^{-3}	9.1	(mg/L)
;	))	SO_4^{-2}	4.3	(mg/L)
affinic hyarbonirbons			4.0	(mg/L)
butyl phose e			50	(mg/L)
suspendedls ds			83	(mg/L)
beta/gam			<225	(pCi/mL)
alpha			<5	(pCi/mL)

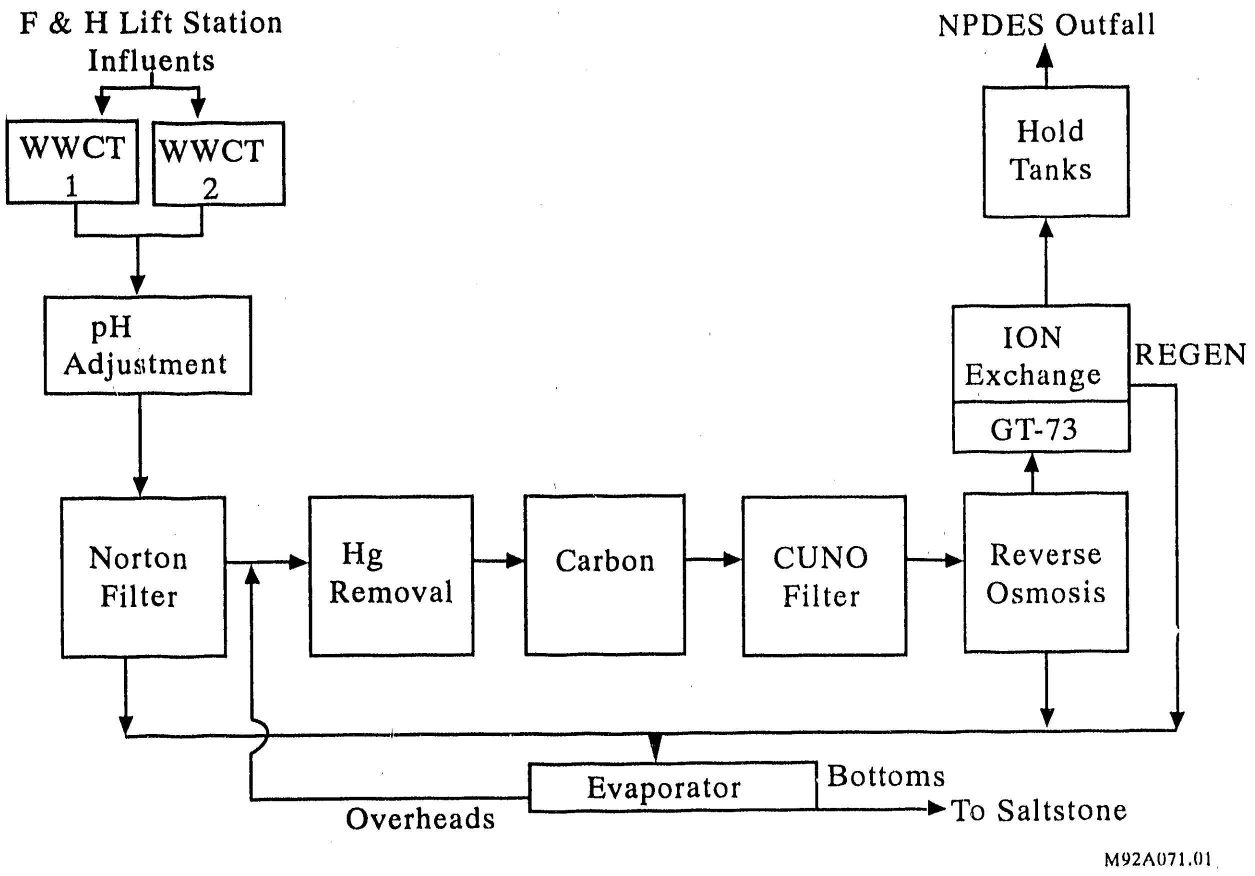

Figure 1. F/H-Area Effluent Treatment Facility Process Flowsheet

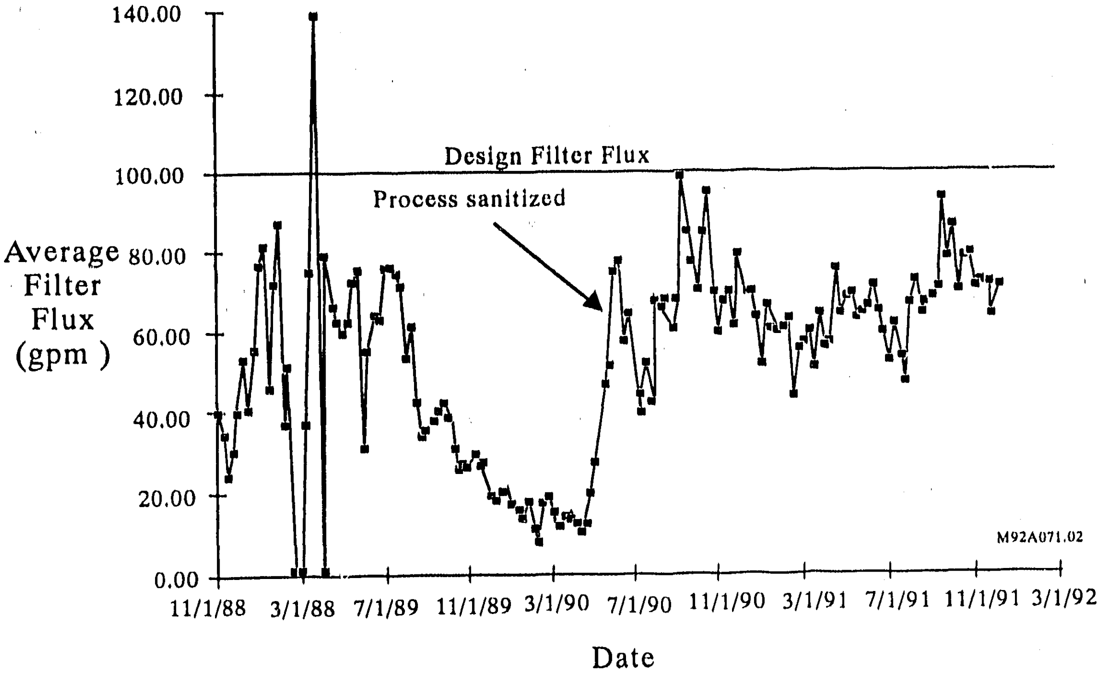

Figure 2. ETF Filter Performance History

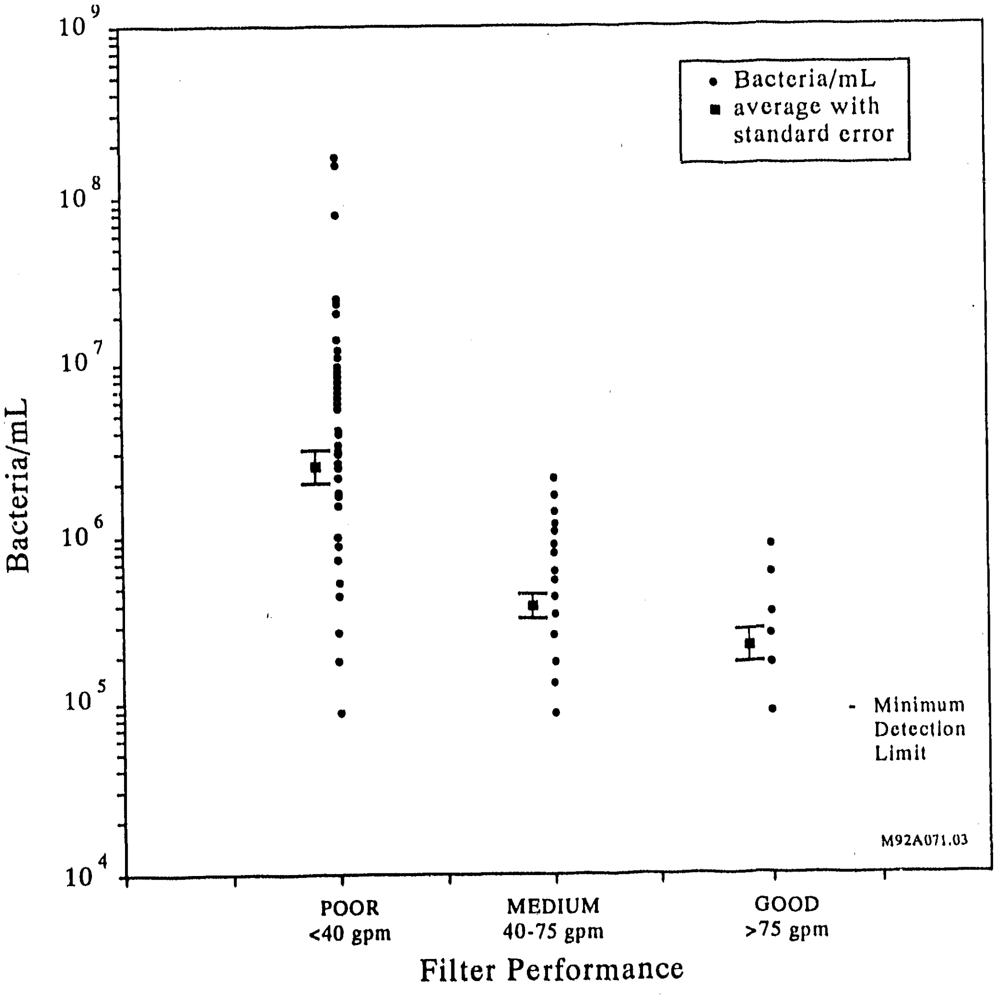

Figure 3. ETF Bacteria vs Filter Performance

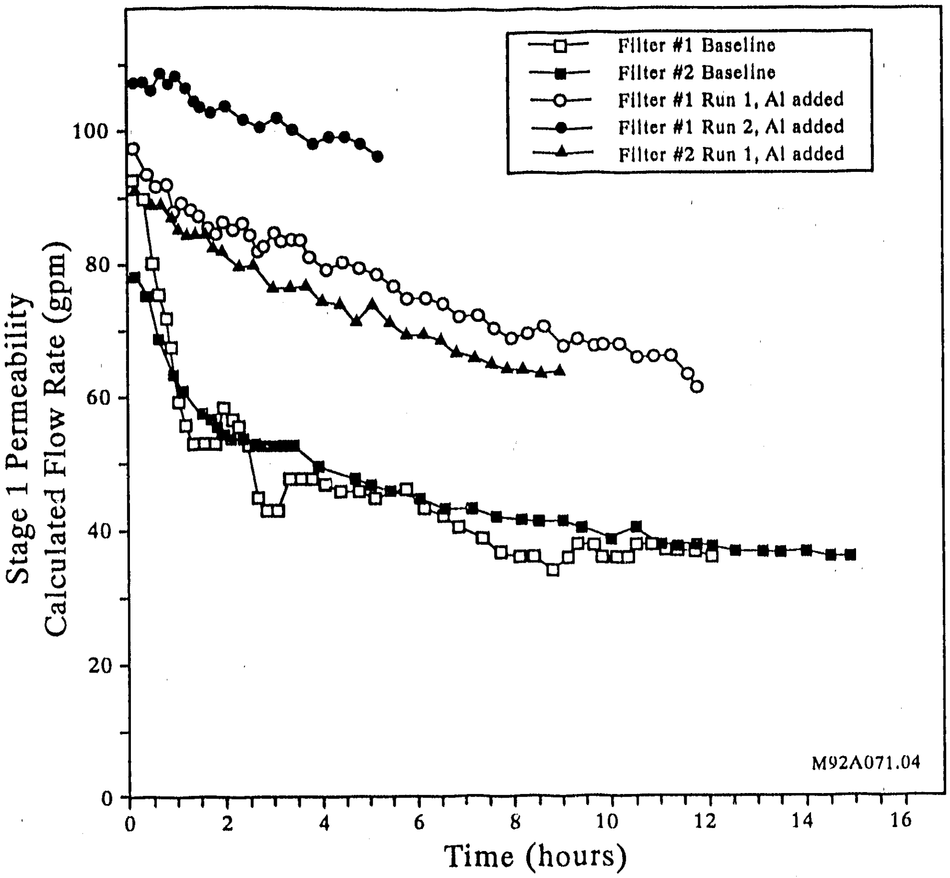

Figure 4. ETF Aluminum Nitrate Test

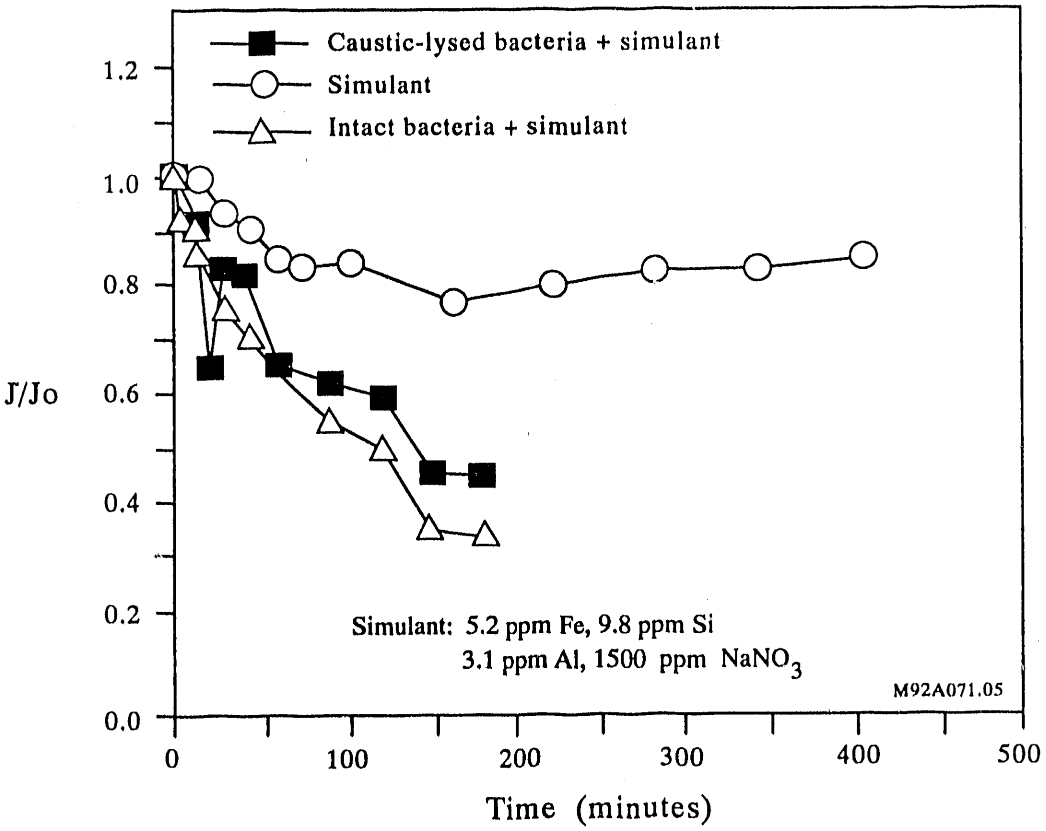

Figure 5. Flux vs Time Caustic-Lysed Bacteria

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