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IMPROVEMENT OF REVERSE OSMOSIS FOR MUNICIPAL WASTEWATER RECLAMATION THROUGH PRETREATMENT

by

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ABSTRACT

Six years of operation of a 10-gallon-per-minute tube-style reverse osmosis pilot plant for reclaiming municipal wastewater are described. The evolutionary development of cleaning and pretreatment techniques, which improved recovery from an average of less than 30 percent to over 70 percent are also described. A cleaning technique was developed which restored membrane flux to virtually 100 percent of new membrane flux. Pretreatment techniques evaluated included coagulation/ filtration of trickling filter and activated sludge plant effluent, using both organic and inorganic coagulants. The activated sludge plant effluent provided much better feed water, with only one-third the fouling tendency of trickling filter effluent. In all cases, aluminum sulfate was the poorest coagulant in reducing the fouling tendency of the feed water, although it sometimes produced the lowest turbidity feed water. In all cases, the greatest level of pretreatment provided the lowest operating cost and least energy consumption.

INTRODUCTION

In an effort to develop future water resources for the state of California, the California Department of Water Resources and others have funded a series of projects to develop technology to reclaim wastewater discharges. The development of additional water resources from wastewaters is one method of meeting the future water needs while reducing wastewater discharge problems. Previous projects have been described by Antoniuk and McCutchan (1973) and Speight and McCutchan (1979) for irrigation drainage wastewaters; by Argo and Moutes (1979), Wojcik, Lopez, and McCutchan (1980), and Stenstrom et al (1982) for domestic wastewaters; and by Johnson and Loeb (1969), Johnson, McCutchan, and Bennion (1969) for saline groundwaters. Other work has been performed in conjunction with the California Department of Water Resources, and the review by Davis, et al (1980) should be consulted for additional information.

This paper describes the two phases of operation of the Las Gallinas pilot plant located at the Las Gallinas Valley Sanitary District, north of San Rafael, in Marin County, California. The objective of this study was to investigate pretreatment techniques and their effect on system performance and cost. The first phase investigated the economics and system design of a pilot scale tubular reverse osmosis plant treating coagulated and filtered trickling filter effluent. Results of this work, through extensive investigation of coagulation filtration techniques, including coagulation by organic polymers, ferric chloride, and alum (aluminum hydroxide), showed that pretreatment would significantly reduce total costs. It was concluded from this work that total operating cost could be reduced by 10 percent by employing optimum coagulation-filtration pretreatment, as compared to untreated trickling filter effluent. Additionally, the fouling materials removed from the RO membranes appeared to be organic in nature, indicating that additional improvements in biological pretreatment would be beneficial.

In the second phase of research, improved pretreatment was provided by a pilot scale activated sludge plant, followed by various filtration/coagulation alternatives. The best alternative would reduce operating cost by 35 percent as compared to using untreated trickling filter as feed water.

EXPERIMENTAL APPARATUS

The reverse osmosis apparatus used in this study was very similar to the units used in earlier investigations conducted by UCLA researchers (Johnson and Loeb, 1966; Johnson et al 1969; Speight and McCutchan, 1979). The unit is very similar to the original design by Loeb and Sourirajan (1960, 1962). Table I lists the specifications for the unit.

TABLE I

REVERSE OSMOSIS UNIT SPECIFICATIONS

Parameter	Value
Membrane Configuration	Tubular
Internal Diameter	0.88 inch
Membrane Material	Cellulose Acetate
Annealing Temperatures	88 to 90°C
Number of Membranes	160
Operating Pressure	600 psig
Feed Rate	6.4 gpm (Phase I)
Feed Rate	3.7 gpm (Phase II)

The tube-style membranes have been used throughout the UCLA research projects, in part because they allow for membrane development and testing without requiring extensive equipment and facilities. The tube-style membranes are particularly useful in reclamation studies since they can be used with the largest range of feed water qualities and can be easily cleaned. Figure 1 shows a cross section of the membrane configuration used at Las Gallinas. Each membrane is 0.88 inch in diameter and 10 feet long, providing a total surface area of 2.24 ft^2. The entire RO unit contained 160 membranes, for a total area of 358 ft^2.

Pilot Plant Description

The pilot plant was located at the Las Gallinas Valley Sanitary District north of San Rafael, California. The District operates a secondary treatment plant composed of primary sedimentation, two-stage trickling filters, and secondary clarification. The flow rate to the plant ranges from the average value of 1.5 mgd (0.065 m^3/s) to upwards of 6 mgd (0.26 m^3/s) in wet weather. The trickling filters are loaded at a rate of 11 mgd/acre (1.17×10^{-4} m^3/m^2-s) and 84 lb of biochemical oxygen demand (BOD^5) per 1000 cubic feet of filter media (1.35 kg BOD_5/m^3). This loading is considered to be a high loading rate according to current design standards and, at this loading rate, the filters are expected to produce effluent BOD_5, ranging from 12 to 25 mg/l (Reynolds, 1982; Metcalf & Eddy, 1979). This effluent BOD_5 concentration compares to 5 to 15 mg/l to be expected from a well designed and operated activated sludge plant (Metcalf & Eddy, 1979). The Las Gallinas plant showed seasonal fluctuations in treatment efficiency. In the winter, the effluent was visibly poorer than in summer, with turbidities exceeding 20 NTU on numerous occasions. The decrease in effluent quality in winter can be primarily attributed to the increase in flow, although the filters would also perform more poorly at cooler wastewater temperature. The entire RO unit and pretreatment facilities, excluding the activated sludge plant, are shown in Figure 2.

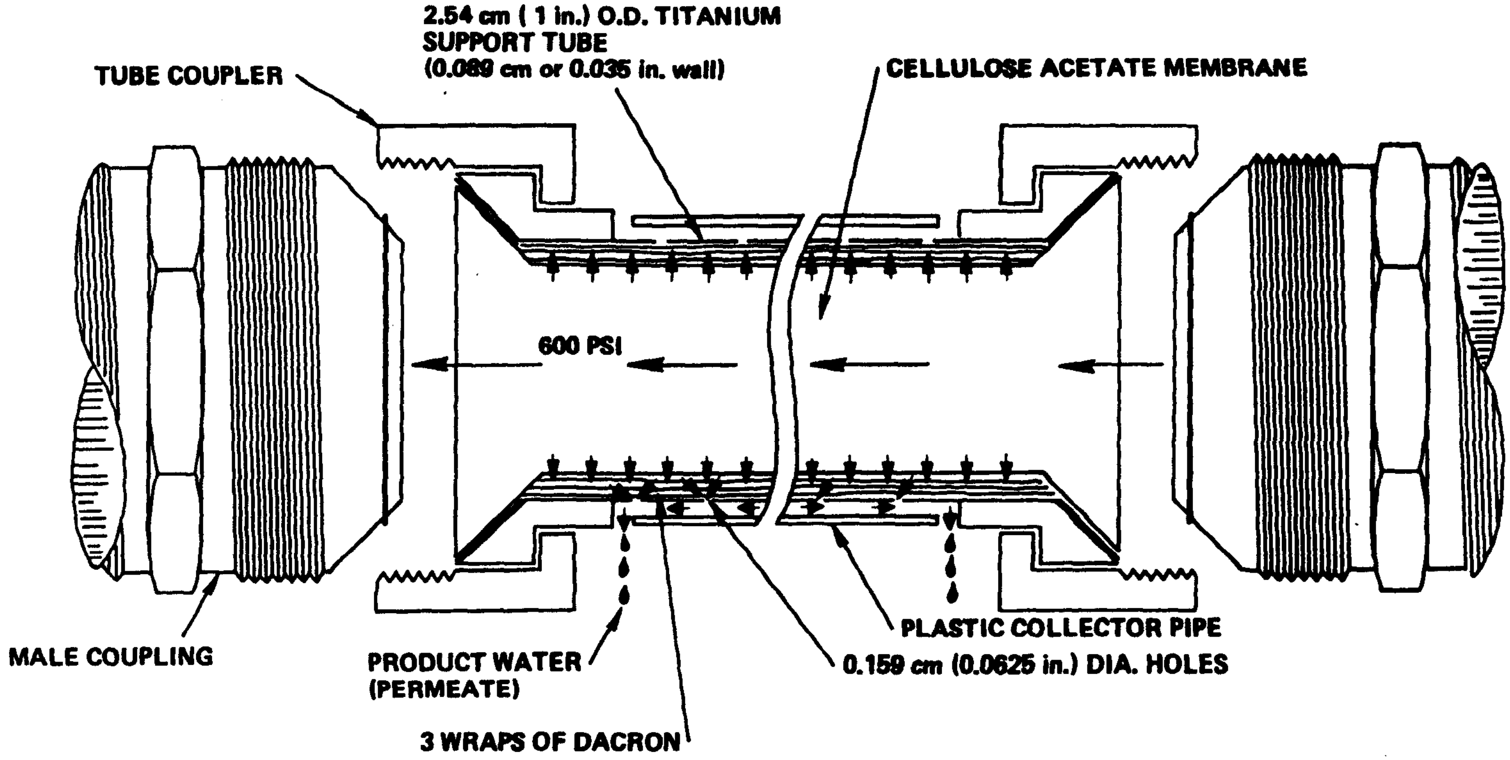
The activated sludge plant was designed from a "package plant" available from the Clow Corporation. The plant was designed to be a self-contained unit, which can be trucked to a site, unloaded, hooked up to utilities, and placed in service. Package plants often serve small subdivisions prior to the construction of sewers, or remote locations such as National Parks service facilities. The unit was constructed to provide dispersed flow operation ("plug flow," in the parlance of treatment plant operators). Provisions were made for tapering the aeration rate and for step feed operation (Torpey, 1952).

The secondary clarifier was constructed as a conical section welded to the rectangular aeration tank. Return sludge was pumped by an air lift pump. No rake was provided and skimming was performed by an open pipe skimmer located at the clarifier surface near the effluent weirs. Figure 3 shows the activated sludge plant. The specifications and operating parameters for the plant are described in Table II.

Chronology of RO Plant Operation

The RO pilot plant was placed in operation in April of 1976, treating trickling filter influent, which was filtered through a 30-inch-diameter multi-media (sand and coal) filter. This filter, in addition to providing feed water for the RO unit, supplied the Marin Municipal Water District (MMWD) with water for its reclamation activities. The RO unit operated on filtered trickling filter effluent from the Marin filter until May of 1979, when a smaller, 24-inch-diameter, mixed-media filter was installed and dedicated to pretreatment of RO feed water.

This initial period, from April of 1976 to June of 1979, was dedicated to the development of membrane cleaning techniques and endurance testing of the RO membranes and equipment. The original cleaning technique was restricted to sponge ball cleaning without chemical cleaning agents. (Sponge ball cleaning was developed earlier by McCutchan and co-workers, and uses a sphere of flexible plastic or rubber foam that can be compressed and forced through the tubular membranes with brine flow.) During cleaning, the unit was always depressurized and flushed with tap water or RO product water (later containing cleaning chemicals). Beginning in April of 1977, a two-hour enzyme detergent flush was initiated. In June of 1977, the detergent flush was stopped and a citric acid flush was begun. Combinations of cleaning techniques were evaluated until March of 1978, when a final cleaning procedure, consisting of one-hour flushes with citric acid and detergent,



REVERSE OSMOSIS TUBULAR SECTION

Figure 1 MEMBRANE CROSS SECTION

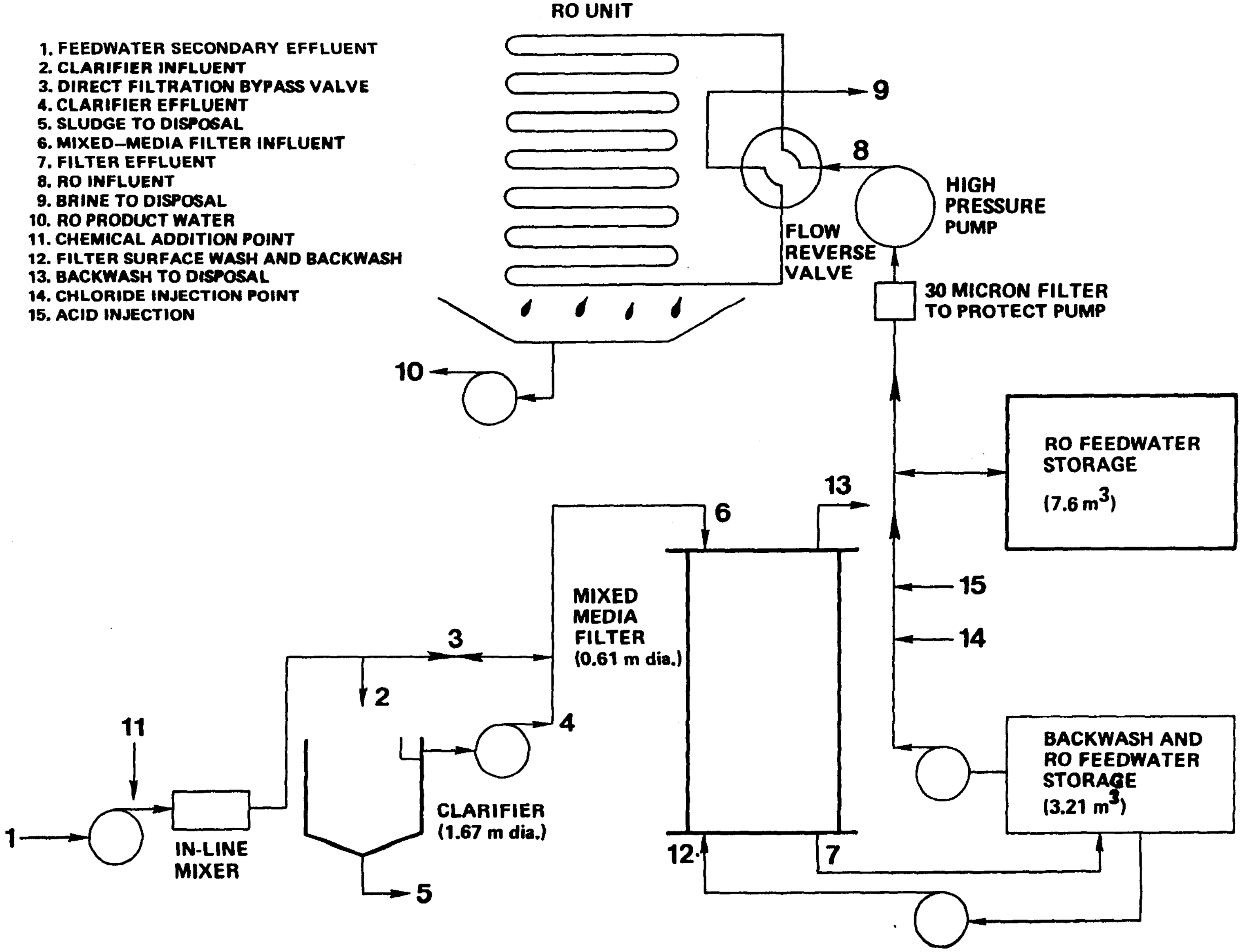


Figure 2 RO PILOT PLANT AND PRETREATMENT FACILITIES

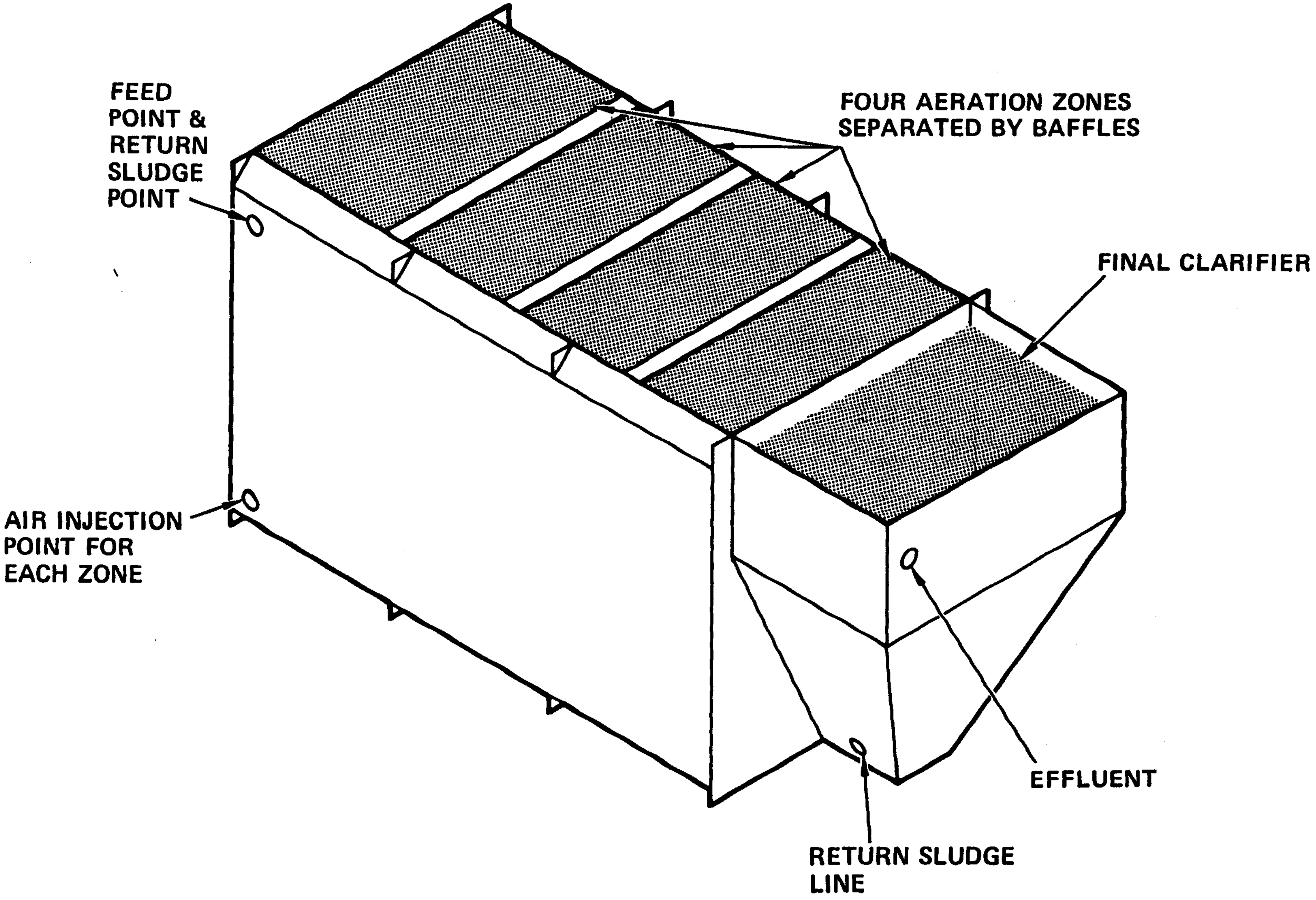


Figure 3 ACTIVATED SLUDGE PLANT

TABLE II

SPECIFICATIONS AND OPERATING PARAMETERS FOR THE ACTIVATED SLUDGE PLANT

Parameter	Value
Aeration Section	
Depth	9 ft (2.75 m)
Length	14 ft (4.25 m)
Width	8 ft (2.43 m)
Volume	7500 gal (28.4 m^3)
Clarifier Section	
Depth	9 ft (2.75 m)
Length	4.5 ft (1.37 m)
Width	8 ft (2.43 m)
Volume	1450 gal (5.5 m^3)
Surface Area	36 ft^2 (13.3 m^2)
Hydraulic Retention Time at 12 gpm	10.4 hours
Overflow rate at 12 gpm	480 gal/ft^2 day (4.9 m/day)
Mean Cell Retention Time	5 days

*12 gpm = 0.045 m^3/min

followed by sponge ball cleaning, was developed. Table III summarizes the final cleaning procedure.

In March of 1978, chlorination of RO feed water was begun and, in May of 1978, pH control of feed water was started. Operation continued in this fashion until June of 1979, when the improved pretreatment facilities were placed in service.

From June until July 1979, feed water was pretreated using direct filtration with a cationic organic polymer (Nalco 7134). In July of 1979, a 5.5 ft (1.7 m) diameter clarifier was installed and inorganic coagulants were used. The mixed-media filter was operated at 3.2 gpm/ft^2 (2.17 1/m^2 s) filtration rate and backwashed at 18 to 20 gpm/ft^2 (10.2-13.6 1/m^2) after a two-minute surface wash. Backwashing was performed automatically on a timed cycle. Usually, backwashes were performed every 12 hours. The filter was operated at the 3.2 gpm/ft^2 (2.17 1/m^2) rate independently of the RO feed water rate in order to provide uniform operation. Excess feed water was discharged with the Las Gallinas Valley Sanitary District Effluent. The filter medium used was a commercially available medium (Neptune-Microfloc) consisting of 1.0- to 1.2-mm-size distributions of coal, a 0.42- to 0.55-mm-size distribution of silica sand, and a 0.2- to 0.3-mm-size distribution of garnet sand.

TABLE III

FINAL MEMBRANE CLEANING PROCEDURE

Operation	Procedure
Citric Acid Flush	0.55 lb (250 grams) of citric acid is added to 50 gallons (190 1) of tap water or RO product water at ambient temperature. This solution is circulated through the RO unit at approximately 5 gpm (0.315 1/s) for one hour.
Enzyme Detergent Flush	1.10 lb (500 grams) of a commercially available enzyme detergent is added to 50 gallons (190 1) of tap water at ambient temperature and circulated through the RO unit as before for one hour.
Sponge Ball Cleaning	After completion of chemical cleaning, ten 1-1/2 inch (3.8 cm) sponge balls are introduced into the RO feed at approximately one-minute intervals, and are allowed to pass through the unit at approximately 2.7 ft/s (0.52 m/s).
Sponge Ball Cleaning	After approximately 70 hours of operation, the unit is depressurized and the sponge ball cleaning is repeated.

The clarifier was operated at a flow rate of 10 gpm (0.63 1/s) also, giving an overflow rate of 610 gal/ft^2 day (24.8 m^3/m^2 day). Sludge was manually withdrawn on a regular basis.

The activated sludge plant was delivered to the Las Gallinas site in January of 1981 and placed in service in April of 1981. The plant was operated for two months to reach steady state. After reaching steady state in June of 1981, the entire pilot facility was placed in service, with the activated sludge plant providing feed water for the RO unit. After a few hours of operation, it was determined that the RO membranes were removing only 5 to 10 percent of the influent total dissolved solids concentration (as measured by specific conductivity) and that the recovery rate was unusually high.

After examination of the membranes, it was determined that they had deteriorated during storage. They were stored in the RO unit under approximately 50 psig of tap water pressure. The actual mode of deterioration remains unknown, but an analysis of a sample membrane indicated that the deterioration was consistent with hydrolysis and oxidation by chlorine. The destruction of the membranes was surprising since the replacement membranes were normally filled with tap water during storage. Also, the membranes in normal operation are exposed to as much as 2.0 mg/l total chlorine residual. It was hypothesized that the destruction of the membranes occurred because of the chlorine contained in the Marin tap water. The Marin tap water usually contained a residual of less than 0.5 mg/l chlorine, but the residual was always in the form of free chlorine (HOCl or OCl^-). Since the Las Gallinas trickling filter never nitrified during the entire study, the residual chlorine in the RO feed water was always a combined residual (primarily monochloramine), known to affect cellulose acetate membranes less severely than free chlorine (Zachariah, 1982). In retrospect, it is now known that the membranes should have been stored in the absence of free chlorine. One method of preserving the membranes would have been to inject ammonia in the tap water when the membranes were being preserved.

It is also possible that the membranes were destroyed by bacterial action; however, this is considered less likely by the author since the tap water was essentially free of bacteria and biological compounds. Also, those membranes failed that had never been exposed to wastewater or used previously.

It was necessary to recast all the membranes before continuing operation. Recasting was performed at DWR's Firebaugh, CA facility (where all of the membranes in this project, and many of the others cited, were cast), and reinstallation was completed at the beginning of January 1982. The casting procedure and facilities have recently been described by the Department of Water Resources (1983). During the period from June 1981 to January 1982, the activated sludge plant remained in operation, but no data were collected.

The activated sludge plant was operated in the "conventional" mode (Metcalf & Eddy, 1979) during the entire study period. The feed water was pumped to the activated sludge plant from the launder of the Las Gallinas east primary clarifier through a 1-1/2-inch PVC pipe line, using a submersible sump pump. Flow rate was monitored manually and ranged from 8 to 13 gpm. The large fluctuation in flow rate was caused by sliming of the PVC line. Weekly cleaning was instituted by injecting several hundred milliliters of a chlorine bleach, followed by flushing with a 3-inch sponge ball. The flush was bypassed directly to a return sewer in order to prevent the chlorine from entering the activated sludge plant. The weekly cleaning helped control flow rate changes, but some variation still occurred.

The skimming device in the secondary clarifier was manually set to flow at approximately 0.25 gpm, but skimming was sporadic. Occasionally, large quantities of scum would form and partially block the skimmer. At other times, wind velocities of changes in flow rate would cause the skimmer to remove less than the desired 0.25 gpm. When the skimmer malfunctioned, scum was carried over to the downstream filter operation, causing increase headloss and premature breakthrough of turbidity.

After repeated attempts to improve skimming, it was decided to operate the 5.5-foot-diameter clarifier, which would function as a second skimming device. After placing this clarifier in service for skimming, no further problems with scum entering the downstream operations occurred. The clarifier removed very few suspended solids and rarely accumulated significant quantities of sludge, although it was periodically drained. The clarifier provided insurance against solids carry-over. In an actual plant, this problem would not occur, since mechanized skimmers would be provided, and operational intervention would be expected in the event of their failure. Therefore, the clarifier is not included in later economic analysis where direct filtration following activated sludge treatment was used.

Sludge was wasted from the activated sludge plant directly from the mixed-liquor, using a Moyno pump with a variable speed dc motor. A sludge age of four to six days was maintained. In this manner, suspended solids determinations were not required to maintain sludge age control. Recycle sludge flow rate was maintained at approximately 8 gpm and variations occurred because of occasional clogging of the return line (since gravity flow was used after an air lift pump).

Table IV summarizes the period of operation and timing of significant events. Throughout the entire period of operation, the units were maintained almost without day-to-day manual supervision. Perhaps 0.5 to 1.5 hours per day were spent on maintenance and operation, with the exception of membrane cleaning, and most of this time was spent logging data and mixing coagulants.

TABLE IV

CHRONOLOGICAL SUMMARY OF PILOT PLANT OPERATION

Date	Hour	Event	Comment
4/27/76	0	Pilot plant started up on trickling filter effluent after multi-media filtration.	Weekly sponge ball cleaning without chemicals.
4/18/77	8,500	Cleaning procedure changed by the addition of two-hour enzyme detergent flush.	Various concentrations of detergent (up to 2.1 g/l) were used for flushing.
6/20/77	10,000	Citric acid substituted for enzyme detergent.	Concentrations between 0.04 and 0.53 g/l were used.
9/26/77	12,400	Returned to enzyme detergent.	Concentrations between 1.05 and 0.32 g/l were used.
1/1/78	14,700	Final cleaning procedure developed, using one-hour citric acid flush, followed by one-hour enzyme detergent flush, followed by sponge ball cleaning.	0.66 g/l citric acid concentration and 1.32 g/l detergent used for flush.
3/23/78	16,700	Chlorination of multi-media filter effluent begun.	Chlorine residual ranged from 0.5 to 6.0 mg/l, averaging 2.0 mg/l.
5/15/78	18,000	Influent pH control initiated by addition of sulfuric acid.	Set point at pH = 5.5.
8/1/78	19,800	Automatic sponge ball cleaning begun.	Cleaning frequency set at six hours.
6/1/79	27,100	Mixed-media filter cationic polymer coagulation initiated.	Dosage set by Zeta potential measurements.
7/6/79	28,000	Clarifier installed and operation with various coagulants and modes until shut down	Optimal concentrations of $FeCl_3$, $Al_2(SO_4)_3$ evaluated
1/7/80	32,400	Unit shut down.	Membranes stored under pressurized tap water.
4/1/81	43,200	Activated Sludge plant started up.	
6/1/81	44,600	RO unit started up and shut down.	Membranes destroyed
1/22/82	49,400	RO restarted using activated sludge plant as feed water.	Operated with various coagulants until shut down.
6/23/82	53,000	Unit shut down and disassembled.	

Analytical Measurements

Most of the analytical work was performed on site using the existing laboratory facilities. Turbidities were measured with a Turner Designs Model 40-005 turbidity meter. Flow rates were usually measured by clocking flows into vessels of known volume. Extensive analyses of the influent and effluent water quality parameters were performed periodically by the Department of Water Resources Laboratories, using Standard Methods (1975) techniques. Total dissolved solids (TDS) were always measured using a specific conductivity meter, but were measured gravimetrically by the DWR laboratory.

Flux Decline Tests

In order to evaluate the effectiveness of various pretreatment techniques in preventing flux decline, 24-hour flux decline tests with close monitoring of flux and product TDS were performed. These tests were conducted during three periods of three to four tests each during the first phase of this project, and over the last week of operation during the second phase. Tests were purposefully performed in clusters in order to prevent the effects of changing influent composition and temperature from obscuring the effects of pretreatment alternatives. The general procedure for performing the test is summarized as follows:

1. To prepare for the test, injection of the coagulant to be evaluated was begun at the clarifier influent pump discharge. In the event that direct filtration was being evaluated, the clarifier was bypassed. The pretreatment system was allowed to operate for several hours in order to come to steady state before turning on the RO plant.

2. The multi-media filter was backwashed, and the pretreatment system was turned on.

3. The RO unit was chemically cleaned with a one-hour citric acid flush, followed by a one-hour enzyme detergent flush, followed by cleaning with ten oversized sponge balls, introduced at one-minute intervals.

4. During RO cleaning, the 2,000-gallon feed tank was drained, flushed with water from the pretreatment system (now operating under test conditions), and allowed to fill.

5. The RO unit was started and adjusted to a feed rate of 3.7 gpm (6.4 gpm in the first phase) and a pressure of 600 psig. Data collection was initiated 30 minutes after start-up.

6. Brine and product flow rates were periodically determined by timing 30 to 60 seconds of flow into 0.264-gallon (1000 ml) graduated cylinders and recording the results in milliliters per minute and gallons per minute. The feed flow was calculated by summing the brine and product flows. TDS was measured and recorded; also recorded were turbidities, power usage, operating pressure, and pH.

7. The measurements were repeated at hourly intervals for the first few hours of the test (usually seven hours) and then repeated again the next morning.

8. After the final measurements, the pretreatment system was shut down and preparations were begun for another 24-hour test.

In the first phase, using treated trickling filter effluent, 24 hours were sufficient to determine flux decline rates. In the second phase, using activated sludge plant effluent, the flux decline tests were conducted over 48 hours. This increase was necessary because of reduced fouling rate of the activated sludge plant effluent.

EXPERIMENTAL RESULTS

Flux Decline and the Effects of Cleaning

The earliest results with the RO unit were disappointing because very low recovery rates were obtained. The recovery averaged about 25 percent with fluxes in the range of 4.5 to 5.0 gal/ft^2 day (gsfd) or 7.6 to 8.5 l/m^2 hr. The earliest use of the sponge ball was effective in restoring the flux to 9 to 10 gsfd after cleaning. After about 8,000 hours of operation, the flux before cleaning decreased to approximately 3.5 gsfd, while the flux after cleaning was restored to only 4.2 to 4.5 gsfd. The deterioration was caused by the precipitation of insoluble salts on the membrane surface, which were not removed by the mechanical cleaning of the sponge balls.

The use of the enzyme detergent partially restored the membrane fluxes, but results were still disappointing. Starting in April of 1977, the fluxes after detergent and sponge ball cleaning gradually increased from 4 to 4.5 gsfd, to a maximum of 5 gsfd. In June of 1977, the first citric acid cleaning was performed, which restored membrane flux to 12.5 gsfd. This flux, after cleaning, was maintained until the end of September when flushing only with the detergent was begun again. The flux, after cleaning, gradually declined and, by December of 1977, had declined to the previous levels of 4 to 4.5 gsfd. Beginning in March of 1978, the final cleaning procedure, shown previously in Table III, was consistently used, and flux after cleaning stabilized to 12.5 gsfd. The results in improvements in cleaning techniques can be seen in Figure 4, which shows the entire period of investigation for Phase I.

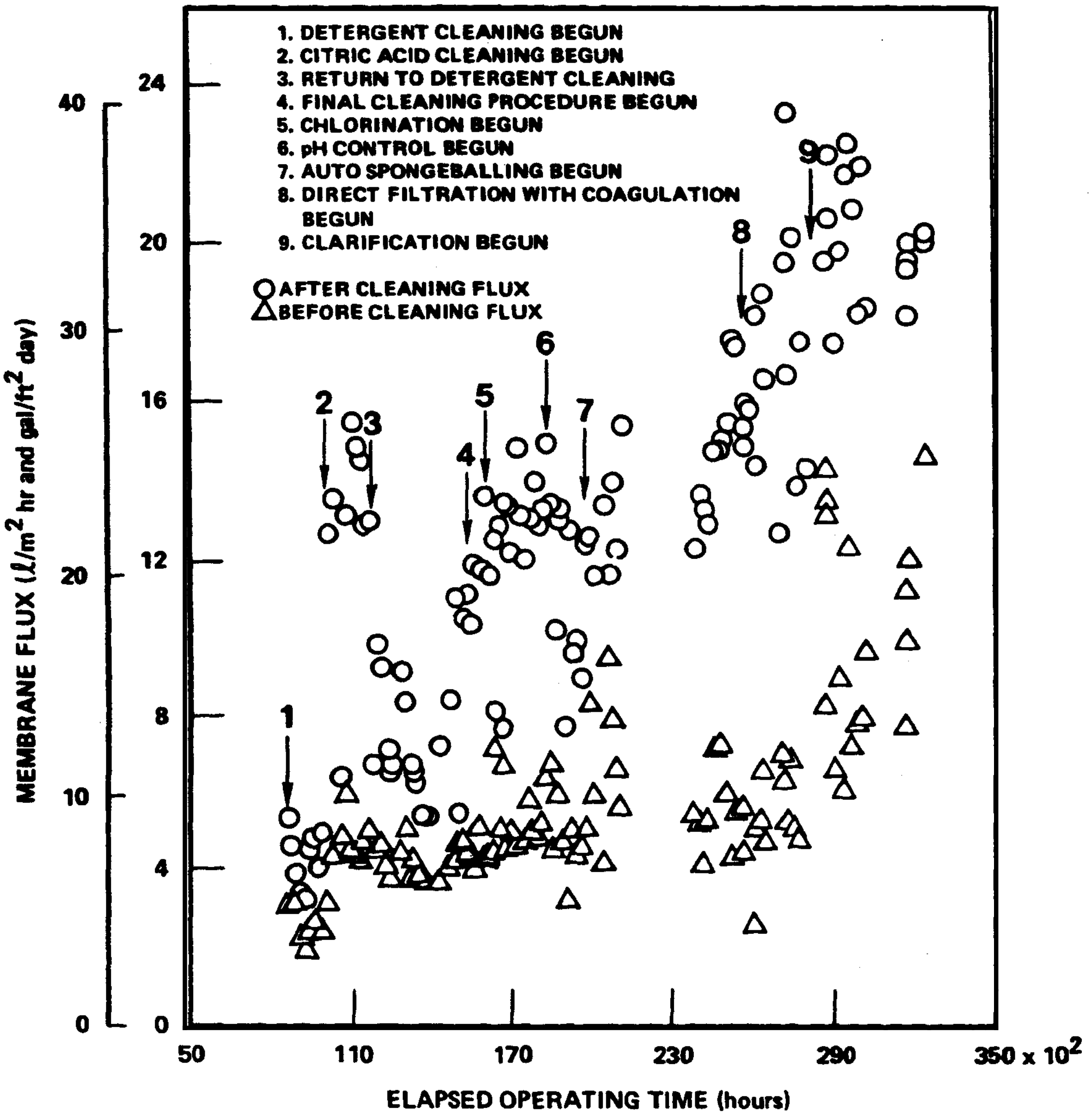


Figure 4 MEMBRANE FLUXES BEFORE AND AFTER CLEANING AS A FUNCTION OF TIME

The increases in flux due to improved pretreatment are obvious. The increases in fluxes after January 1978 were caused by improved pretreatment techniques, rather than additional membrane-cleaning techniques. The cleaning technique provides essentially complete membrane cleaning. The cleaning technique remained the same for the second phase of study.

Feed Water Quality

Figure 5 shows the turbidities as a function of time for the activated sludge plant effluent, clarifier effluent, and mixed-media filter effluent over the first 100 days of the Phase II period. The filter effluent averaged well below 2 NTU, which is significantly less than in Phase I using trickling filter effluent, when mixed-media filter effluent turbidities ranged from 2 to 5, and were seldom less than 2.0.

It can be observed from Figure 5 that the clarifier effluent turbidity is little different than the activated sludge plant effluent. It was noted earlier that the clarifier was used primarily as a second scum control device, and that, in the design analysis, it was neglected. This figure supports the assumption that the clarifier would not be required in a full-scale design where mechanized skimming facilities would be available.

On April 5, 1982 and at other times during the study, samples of feed water and product water were collected and analyzed by the DWR Sacramento laboratory. Table V shows the results of these analyses, along with two sets of analyses from Phase I. The results from Phase I are very similar to the results obtained in Phase II. The feed water varies somewhat from Phase I, as expected. The primary difference in feed water properties is the nitrogen compounds and its forms. The nitrogen was almost 100 percent ammonia and organic nitrogen in Phase I while, in Phase II, the activated sludge plant provided partial nitrification, reducing the ammonia concentration while increasing the nitrite and nitrate concentrations. This difference is also reflected in the product water, since ammonia removal with cellulose acetate membranes is poorer than nitrate and nitrite removal. In Phase I, the total nitrogen of the product water was in the range of 4 to 7 mg-N/l while, in Phase II, it was less than 2.0 mg-N/l. The TDS of the April 5th analysis is somewhat lower than the average for the Phase II period. This resulted from infiltration of rain water, which occurred around April 5th. High wet weather flows are typical at the Las Gallinas Valley Sanitary District. The total organic carbon (TOC) from Phase II is higher than Phase I, which was surprising, and may have occurred because a different analyzer was used in Phase II.

Flux Decline and the Effects of Pretreatment

Improvements made in recovery and flux maintenance after January of 1978 were caused largely by improvements in RO feed water quality. Chlorination of RO feed water was begun in March of 1978, and feed water pH control (pH controlled to approximately 5.5 with sulfuric acid) was begun in May of 1978. Both of these changes resulted in small increases in flux maintenance. The pH control improved flux maintenance because of the increased solubility of calcium sulfates and carbonates at the reduced pH, while chlorination prevented the growth of fouling slimes on the membranes. The actual purpose of pH control was to reduce membrane hydrolysis, but it also had this additional benefit.

The installation of the auto-sponge ball cleaning device in August of 1978 coincides with increases in before-cleaning fluxes to as high as 8 gsfd. Unfortunately, the high before-cleaning fluxes declined to the level of 5 to 7 gsfd during the period of October 1978 to May 1979. No reason for this decline was determined.

The use of chemical coagulation and clarification had very large effects on both before- and after-cleaning fluxes. Direct filtration with a cationic polymer, which was begun on May 31, 1979, coincides with increasing trends in both before-cleaning and after-cleaning fluxes. The before-cleaning fluxes increased to maximum values of about 14 and 25 gsfd, respectively, during the final periods of Phase I, when the inorganic coagulants were used.

FLUX DECLINE TESTS

During various times in Phase I and at the end of Phase II, a series of flux decline tests was made using various concentrations of ferric chloride, alum, and organic coagulants. During the first phase, the tests were conducted over a 24-hour period while, in the second phase, they were conducted over a 48-hour period. Flux decline was also determined for Marin Municipal Water District tap water, to determine the effects of membrane compaction.

Unfortunately, the flux decline tests performed in Phase II are not directly comparable to those shown in Figure 4 because of the slight differences in membrane characteristics. It was noted previously that the membranes were all replaced beginning in July of 1981 because of deterioration during storage under tap water pressurization. The new membranes were cast using the same procedure as previously used, and cured at 88°C as previously cured; however, the flux and sodium rejection properties of the new membranes were different from the old membranes. The new membranes were much "tighter" than the old

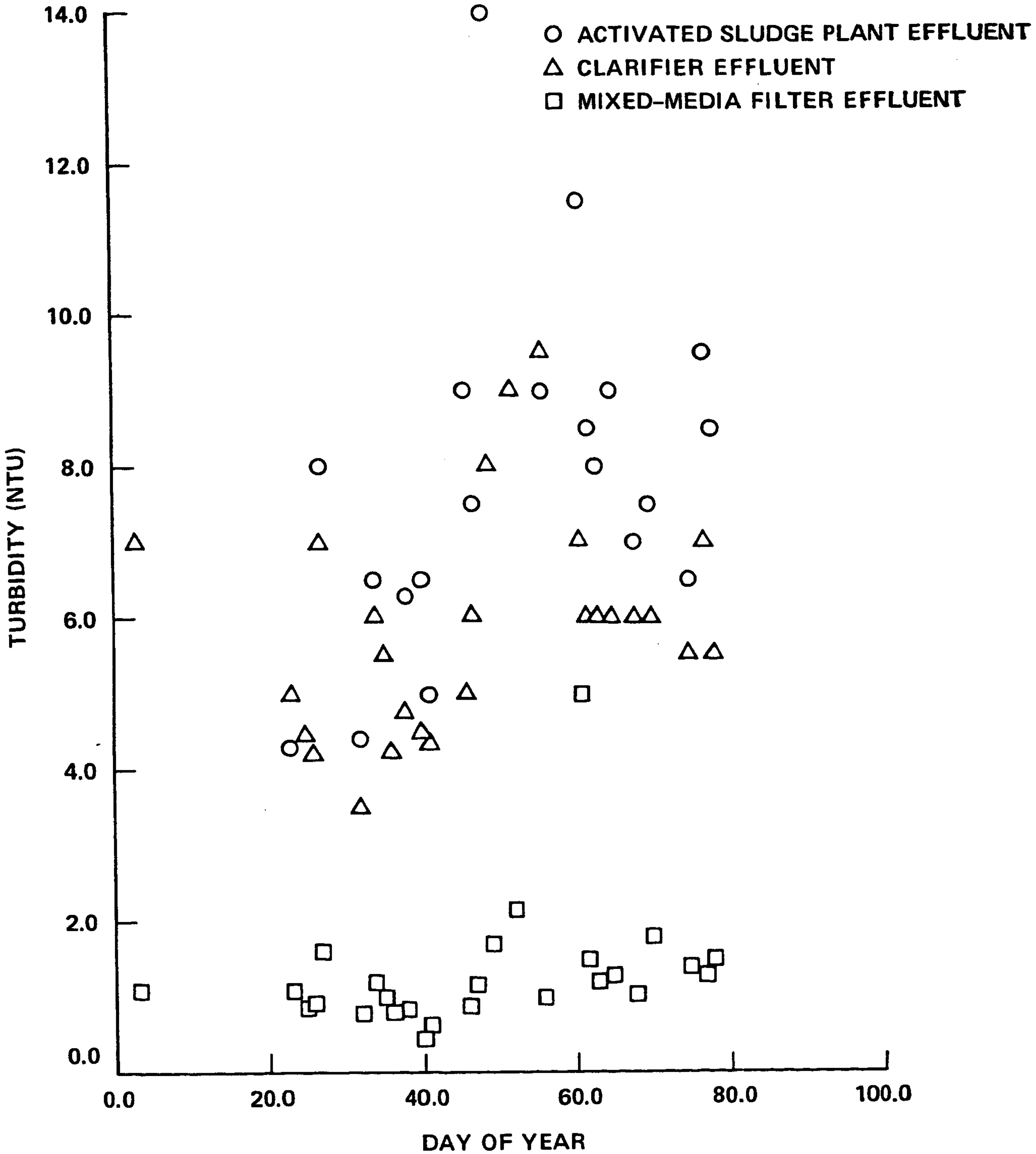


Figure 5 TURBIDITIES AS A FUNCTION OF TIME

TABLE V

CHEMICAL ANALYSIS OF RO FEED WATER AND PRODUCT WATER

Parameter	Feed Water			Product Water		
	3/19/79	9/17/79	4/5/82	3/19/79	9/17/79	4/5/82
Hardness*	216	241	163	13	19	6
Calcium	36	38	29	2	3	2
Magnesium	30	35	22	2	3	1
Sodium	136	218	70	28	57	7
Sulfate	77	251	75	1.5	8.5	1
Chloride	207	351	105	44	91	105
Boron	0.55	0.55	0.20	0.40	0.45	0.20
TDS	671	1090	434	98	203	41
TOC	27	23	36	1.5	1.2	1.9
Total N	26	39	19	4.1	4.7	2.6
Total P	9.3	12	7.0	0.21	0.83	0.05
Iron	0.09	0.36	0.51	0.0	0.08	0.04
Copper	0.01	0.15	0.13	0.0	0.01	0.02
Lead	<0.01	<0.01	0.015	<0.01	<0.01	<0.01

*As $CaCO_3$

In addition, the following constituents were measured, and less than 0.01 mg/l were found in both product and feed water: arsenic, cadmium, chromium, mercury, and nickel.

Note: All values reported in mg/l. Values represent averages of measured water quality before and after chemical cleaning.

membranes. The old membranes removed TDS to an average level of 200 mg/l, while the new membranes initially reduced the TDS to less than 50 mg/l and often less than 20 mg/l. The after-cleaning flux of the old membranes averaged 15 to 20 gsfd, and was sometimes as high as 25 gsfd, while the after-cleaning flux of the new membranes was only 12 to 15 gsfd. This change in membrane properties is consistent with an increase in membrane curing temperature, or may possibly be caused by the newness of the membranes. At the conclusion of Phase I, when most of the 24-hour flux decline tests were performed, the membrane average age was over one year, while the age of the membranes in the Phase II flux decline tests was six months. No resolution of the difference in membrane properties was determined.

In the design analysis performed later to determine the effects of membrane properties on design, a hypothetical condition was created which assumed the existence of a membrane that had the same flux decline properties as the new "tight" membranes, and the same salt rejection properties as the old membranes. This was a conservative assumption because a "looser" membrane should have higher fouling properties when biological materials are present, because of the high flux and high throughput of fouling materials; therefore, flux decline should be higher with the old, "looser" membranes. The economic analysis described later shows that the new, "tight" membranes provide a more economical design; therefore, the question of why the new membranes were different and what their flux decline properties were does not effect the final conclusions of this study.

In comparing the flux decline tests for Phases I and II, some observations can be made. The best case for Phase I, ferric chloride coagulation, showed a decline in flux from 16 gsfd to approximately 11.5 gsfd after 24 hours, or a decline to 72 percent of the after-cleaning flux. For the best case in Phase II, filtration without coagulation, the flux declined from 15 gsfd to 13.6 gsfd, or a decline to 91 percent of the after-cleaning flux. For alum coagulation, the worst case in Phase II, the decline was to 80 percent of the after-cleaning flux.

Small fluctuations in product flow rate were observed in the flux decline tests in Phase II, and were attributed to changing feed water temperature. The actual Las Gallinas effluent temperature fluctuated very little during the day, but the activated sludge plant effluent, when stored in an above-ground tank exposed to sunlight, varied in temperature by several degrees Celsius.

FLUX DECLINE PARAMETER

In order to quantify the flux decline properties of a particular wastewater and at a particular condition, it was necessary to characterize the flux decline mathematically. The method of Thomas et al (1973) was used. Thomas et al plotted the flux declines on log-log paper and found that the slopes were approximately linear, and called the slopes the Flux Decline Coefficient. Flux decline coefficients were calculated for the results from Phases I and II using linear regression of the logs of flux and time, and are shown in Table VI, along with several values reported by Thomas et al (1973). The flux decline parameter (called the "B value" in the computer programs) is a useful method of comparing the fouling tendency of a wastewater. The lower the B value, the lower the fouling tendency. Using the flux decline parameter, the flux decline over time can be simulated for economic and process analysis.

Membrane Replacement

Membranes were periodically replaced as they failed. Failure was monitored by measuring each membrane's flux and TDS. In most cases, failure was obvious because of markedly higher flow. From an analysis of the failed membranes, it was determined that the major failure mechanism was corrosion of the brass end couplings. The very tip of the coupling lip would corrode, which allowed the membrane tip to sever. The average membrane life at the end of Phase I was 10,000 hours. Phase II was too short to determine membrane failure rate.

DESIGN ANALYSIS

In the Phase I work, a design analysis was developed to evaluate the benefits of improved recovery with the costs of increased pretreatment. The details of this analysis are reported by Davis et al (1980), and Stenstrom et al (1982), and will not be discussed in detail here. The techniques were developed in part from previous work by Hatfield (1967); Hatfield and Graves (1970); Fan et al (1970); and McCutchan and Goel (1974). The cost data for the various processes, both capital and operating, compiled by the Oak Ridge National Laboratory (1980) and the EPA (1979), were used to develop the costs of various treatment alternatives. In order to keep the financial calculations current, the costs have been updated through the use of the Engineering New-record cost updates.

The model is based on using the RO product water blended with a specific quantity of RO feed water, to provide water for recycle with a specified water quality. The calculation procedure is to determine the minimum quantity of RO product water for blending with feed water to meet a number of specified water quality standards such as TDS, TOC, turbidity, and Biochemical Oxygen Demand. Pretreatment level and cleaning frequency are considered as variables, while RO operating pressure, membrane characteristics, and initial fluid velocity within the membrane are considered constant. Optimal cleaning frequency was determined and cost was calculated considering downtime and labor for cleaning.

The relative costs of all alternatives, including the four pretreatment alternatives evaluated in Phase I, are presented in Table VII. Also included in Table VII is a hypothetical membrane, described earlier, which was used to evaluate the potential effects of the "tight" membranes used in Phase II. The estimates are made to obtain a blended product water with 500 mg/l TDS. Blending to other water quality objectives can also be obtained. The cost comparisons are made relative to the original 1979 analysis using the trickling filter effluent without additional pretreatment. The cost per thousand gallons for the alternative system, using filtration of trickling filter effluent as feed water, is assigned an arbitrary value of 1.0. The costs of all other alternatives, for 1979 and subsequent years, are made as ratios to the value of 1. This was done because it is difficult to predict accurately the actual costs from scaling up a small pilot plant; however, the relative costs among alternatives should be accurate. The total cost in 1983 dollars for the best alternative, determined from the previously mentioned cost sources, using the Engineering News-record indices for updating is approximately $1.70 per 1000 gallons for a 1 mgd facility. This figure corresponds to the

TABLE VI

FLUX DECLINE PARAMETERS

Flux Decline Coefficient	Feed Water Type	Reference
0.243	Trickling Filter Effluent with Dual Media Filtration	This Study, Phase I
0.202	Trickling Filter Effluent with Alum Coagulation, Clarification, and Mixed-Media Filtration	This Study, Phase I
0.204	Trickling Filter Effluent with Organic Polymer Coagulation, Clarification, and Mixed-Media Filtration	This Study, Phase I
0.146	Trickling Filter Effluent with Ferric Chloride Coagulation, Clarification, and Mixed-Media Filtration	This Study, Phase I
0.0136	Tap Water (TDS = 100 mg/l)	This Study, Phase I
0.059	Activated Sludge Effluent with Ferric Chloride Coagulation, Clarification, and Mixed-Media Filtration	This Study, Phase II
0.037	Activated Sludge Effluent with Direct Mixed-Media Filtration	This Study, Phase II
0.075	Activated Sludge Effluent with Alum Coagulation, Clarification, and Mixed-Media Filtration	This Study, Phase II
0.9	Raw Wastewater	Calculated by Thomas et al (1973) from Feuerstein and Bursztynsky (1970)
0.56	Primary Effluent	Calculated by Thomas et al (1973) from Feuerstein and Bursztynsky (1970)
0.35	Secondary Effluent	Calculated by Thomas et al (1973) from Feuerstein and Bursztynsky (1970)
0.14	Carbon-Treated Secondary Effluent	Calculated by Thomas et al (1973) from Feuerstein and Bursztynsky (1970)

"1.01" entry in the seventh column of Table VII. The cost of other alternatives can be obtained by ratioing this cost to the other table numbers. This absolute cost is considered tentative because of the potential errors of scale-up.

TABLE VII

RELATIVE COST FOR A 1 MGD REVERSE OSMOSIS WASTEWATER RECLAMATION FACILITY

	Type of Process and Pretreatment[a]							
Year	TF FeCl$_3$	TF Organ	TF Alum	TF None	ASP FeCl$_3$	ASP None	ASP Alum	ASP[b] None
1979	0.92	1.06	1.23	1.00	0.72	0.65	0.97	0.76
1980	1.08	1.24	1.42	1.17	0.83	0.76	1.11	0.88
1981	1.20	1.37	1.57	1.30	0.93	0.85	1.23	0.98
1982	1.30	1.49	1.71	1.41	1.01	0.92	1.34	1.07
1983	1.42	1.61	1.86	1.53	1.10	1.01	1.46	1.16

[a] Codes for Process and Pretreatment

TF = Trickling Filter Secondary Treatment

ASP = Activated Sludge Process Secondary Treatment

FeCl$_3$: Coagulation, Sedimentation, and Filtration, Using Ferric Chloride as a Coagulant.

Alum: Coagulation, Sedimentation, and Filtration, Using Alum as a Coagulant.

Organ: Coagulation, Sedimentation, and Filtration, Using an Organic Cationic Polymer as a Coagulant.

None: Filtration with No Coagulation or Sedimentation.

[b] Hypothetical membrane having the same fouling properties as the tight membranes used in the second phase, but having flux and TDS removal properties similar to the membranes used in the first phase.

CONCLUSIONS

The results of a six-year experimental and theoretical analysis of a 10-gpm pilot plant for producing reclaimed water have been presented. These results were applied to the design of a full-scale 1-mgd facility. From this work, the following conclusions are made:

1. Cleaning, using flushes of citric acid, followed by enzyme detergent and sponge ball cleaning, was effective at maintaining membrane flux to essentially the initial flux levels. The citric acid was the major cleaning agent. Enzyme detergent and/or sponge ball cleaning without citric acid were relatively ineffective.

2. The automatic sponge ball cleaning technique appeared to have promise for maintaining membrane flux between chemical cleanings. Further testing is desirable.

3. The major factor contributing to membrane degradation for the type of membranes used in this study was corrosion of the end couplings. The average membrane life during Phase I was 10,000 hours.

4. The activated sludge plant effluent had significantly less tendency to foul the membranes than trickling filter effluent, indicating that a major source of fouling material in Phase I was organic material. The activated sludge plant followed by direct filtration produced feed water that had only one-third the fouling tendencies of the best trickling filter effluent obtained from the Las Gallinas facilities.

5. The greatest level of pretreatment produced the least cost alternative.

6. Alum was always the poorest coagulant, which was probably caused by carryover into the RO unit of aluminum hydroxide, which has minimum solubility at pH = 5.5.

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