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Hydrodynamic Flux Control for Waste Water Application of Hyperfiltration Systems

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HYDRODYNAMIC FLUX CONTROL FOR WASTE WATER APPLICATION OF HYPERFILTRATION SYSTEMS

by

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ABSTRACT

The effect of hydrodynamics on flux decline of cellulose acetate hyperfiltration membranes was studied using primary effluent from the Oak Ridge East Sewage Plant as feed. The system contained multiple, annular-geometry housings which could be operated simultaneously at different velocities with the same feed. Initially, each tube in an annular housing was divided into two sections which could be sampled individually. In the latter part of the program, 3-section tubes were used so that membranes with three different flux capabilities could be exposed simultaneously to three different velocities. The range of fluxes studied was from 2 to 150 gal./ft^2·day and axial velocities ranged from 3 to 30 ft/sec. System pressure was 600 psig.

The results demonstrate that there is a threshold velocity above which flux decline is markedly smaller than at lower velocities. Visual inspection of the membranes after a test indicates that operation above the threshold velocity markedly reduces accumulation of solids during the first 200-300 hours of a test. In scouting studies, addition of a flocculant appeared to markedly reduce the threshold velocity.

During the initial 200-300 hours of those tests with primary sewage effluent as feed and in which the axial velocity was above the threshold velocity, the flux decline parameter, b, [$b = -(\Delta \log \text{flux})/(\Delta \log \text{time})$] was directly proportional to the six-tenths power of the flux and inversely proportional to the square root of the velocity.

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SECTION I

CONCLUSIONS

In studies of the effect of hydrodynamics on flux decline in hyperfiltration of primary sewage effluent with cellulose acetate membranes, convincing evidence was obtained demonstrating that there is a threshold velocity above which flux decline is markedly smaller than at lower velocities. Based on inspection of membranes at the conclusion of tests, it is apparent that operation at velocities above the threshold prevents accumulation of visible amounts of solids on the membrane in the first 200-300 hours of a test. Thus the threshold velocity is that velocity above which deposition of particulates on membrane surface is minimized.

The results of these studies indicate that for primary sewage with no additives, the threshold velocity, u_c, (in ft/sec) is related to the initial flux, J_I, (in gal./ft^2•day) by

$$u_c = (1.3 \pm 0.6)\ J_I^{1/2} \quad .$$

Surprisingly, results obtained at ORNL using untreated Tennessee River water feed and results obtained using Pomona, California secondary effluent as feed are in remarkably good agreement with present threshold velocity results using primary sewage effluent as feed. This suggests that flocculation characteristics of polluted surface waters may be similar. If this proves to be so, then the present results should be applicable to a wide variety of feeds.

In scouting studies in which ~50 mg/ℓ $FeCl_3$ was added to the primary sewage, there was evidence that the value of the coefficient in the threshold velocity equation was reduced from 1.3 to ~0.5. With an initial flux of 50 gal./ft^2•day the threshold velocity was reduced from ~9.5 to ~4 ft/sec. Additional studies are required to substantiate this. If proven true it could have important consequences for the application of hyperfiltration to treatment of municipal sewage effluents.

During the initial 200-300 hours of those tests with primary effluent feed and in which the axial velocity was above the threshold velocity for particulate fouling, the effect of flux, v, and axial velocity, u, on the flux decline parameter, b, ($b = -\Delta \log \text{flux}/\Delta \log \text{time}$) was found to be

$$b \propto \frac{v^{0.6}}{u^{0.5}} \quad .$$

The value of b may also be a function of concentration of foulants although additional tests are required for quantitative evaluation of the relation.

For times longer than 200-300 hours the situation becomes more complicated. Apparently stabilized flux declines can suddenly become markedly worse, even at velocities above the threshold. Based on visual inspection of the membranes after long term tests and on the relative times at which the

increased rate of flux decline began with membranes having different initial fluxes exposed to different axial velocities, it was suspected that this increased rate of flux decline was associated with some sort of nucleation phenomenon, in particular, that solids build-up occurred preferentially at nucleation sites (of unknown origin) leading ultimately to a covering of the membrane by a layer of solids and marked flux decline.

Based on this premise, calcium hypochlorite (~100 mg/ℓ) was added for ~1 hour directly to the primary sewage feed in one test after 620 hours operation. In that test a markedly increased rate of flux decline had begun after 300-400 hours even with membranes operated above the threshold velocity. Almost immediately there was a pronounced flux recovery with those membranes operated above threshold; the recovery was to almost the value based on extrapolation of the flux results for the first 200-300 hours of operation. In a subsequent test, the calcium hypochlorite additions were made for 1 to 2 hours at approximately 250-hour intervals in a test lasting 985 hours. Such treatment prevented the change in the rate of flux decline which began after 300-400 hours in the previous test. Based on observed rejection of the membranes during this test, there was no damage to the membrane during this 985-hour test. However, longer testing times are required to determine whether intermittent treatment with calcium hypochlorite coupled with operation above the threshold velocity is a satisfactory procedure for preventing particulate fouling of hyperfiltration membranes used in treatment of primary sewage effluent.

SECTION II

RECOMMENDATIONS

The present studies clearly demonstrated that deposition of particulates on membrane surface can be minimized by operation above the threshold velocity. However, even when the axial velocity was above the threshold velocity, there continued to be a flux decline. In all probability this flux decline was due to dissolved materials which interacted with the membrane surface in an unknown way.

The primary objective of future studies should be the determination of the nature of this interaction, evaluation of which feed constituents cause the most severe problems and development of procedures for ameliorating the problem.

Secondary objectives should be the development of economic procedures for increasing the floc size, thereby reducing the threshold velocity for particulate fouling, as well as further studies of the nucleation phenomena which occurs after 200-300 hours of operation.

SECTION III

INTRODUCTION

In early studies (1) with a plate and frame module exploring the possible application of hyperfiltration (reverse osmosis) to the treatment of filtered municipal secondary sewage effluent, it was demonstrated that besides removing dissolved salts, the cellulose acetate membranes effectively removed surface active materials, and organic solutes (Table 1). These exploratory studies also indicated that time-dependent flux decline was a major problem when treating municipal sewage effluent by hyperfiltration. Tests were usually of 5 to 6 hours duration. It was found that flux decline was most severe with high initial flux (30 to 60 gal./ft^2•day) membranes. After a few hours operation, flux from these membranes approached values observed with much lower initial flux (10 to 20 gal./ft^2•day) membranes. Even with these lower flux membranes, decreases in flux ranged from 45 to 68% of the initial values in the 5- to 6-hour test period.

In subsequent studies with a spiral wound and tubular modules (2,3) the high organic and inorganic solute rejection obtained in the early tests were again repeated (Table 1). Flux decline was again a problem, particularly with high flux membranes. Among steps taken to ameliorate the problem were carbon pretreatment, chlorination, and a daily air-water cleaning. With a membrane having an initial flux of 5 gal./ft^2•day, a flux decline of ~15% occurred in the first 250 hours; little further decline was noted until after 1100 hours of operation. However even with this pretreatment and daily flushing, a membrane with an initial flux of 17 gal./ft^2•day showed severe flux decline; the flux from this membrane fell below the flux from the 5 gal./ft^2•day membrane after about 800 hours of operation. The test was continued for a total run time of 5000 hours. At that time the rejection of the low flux membrane had deteriorated only by the amount expected due to hydrolysis of the cellulose acetate membrane (3). Thus in this extended run the treated-secondary effluent appeared to have no particular deleterious effect on the membrane.

In exploratory studies with membranes dynamically formed from poly-anionic electrolytes and from sewage constituents, initial fluxes were frequently greater than 100 gal./ft^2•day at 1000 psig pressure; by using high circulation velocities (>30 ft/sec) flux values were frequently over 50 gal./ft^2•day after 400 hours of operation (4,5).

Based on the promising results of these initial studies, several groups made more comprehensive investigations, a substantial portion of which were devoted to attempts to develop procedures for ameliorating the flux decline problem.

Smith, Masse and Miele (7) made pilot plant studies to determine the effect of feed water quality on membrane performance and to examine methods to control membrane fouling. Three different configurations were used in their studies: flat plate, spiral wound and tubular modules. Typical rejections observed in these studies are given in Table 1.

Table 1. Rejection of Various Constituents from Municipal Sewage Plant Effluents by Cellulose Acetate

	AWTR-14 (1965)	Merten and Bray (1966)	Hauk, et al. (1969)	Smith, Masse, Miele (1970)	Nusbaum, Sleigh, Kremen (1970)	Envirogenics (1970)	Conn (1971)
Source Pretreatment	Secondary Filtered	Secondary Carbon	Primary None	Secondary Carbon	Secondary Carbon	Secondary None	Raw Filter Aid
Flux, gfd	~10	~10	33	10-25	13-17	50-60	6-8
Pressure, psi	750	400	1000	400-800	600	600	500
Rejections:							
(a) BOD	98	–	86	–	–	–	–
(b) Organic Carbon	–	90	–	90-95	~90	80-100	–
(c) Inorganic Carbon	–	–	–	–	–	–	–
(d) ABS	98	>97	93	–	–	–	85-87
(e) TDS	97	96	97	93-95	94	89-95	90-93
(f) Cl^-	69	95	–	–	94	60-85	–
(g) NO_3^-	–	~80	50	60-70	54	31-75	–
(h) $PO_4^{\equiv}$	>96	>99	>99	95-99	>99	95-99	96-98
(i) $Ca^{++} + Mg^{++}$	98	>99	–	–	96	–	–
(j) NH_4^+	86	>85	–	80-90	85	80-100	–

With the spiral wound module, a daily 15-minute air-tap water flush with the system in a depressurized condition was successful in controlling excessive pressure drops across the brine side of the spiral wound modules. However, these flushes had little effect on the rate of flux decline. Two other methods were tested, a citric acid flush and a flush with anionic detergent solution. Neither of these procedures were successful in restoring membrane flux.

With the tubular configuration two different runs were made with carbon treated secondary effluent. The initial flux in each run was 20 to 25 gal./ft^2•day. At an axial velocity of 2.0 ft/sec, flux decline was 11% after 300 hours while at an axial velocity of 0.4 ft/sec flux decline was 41% during the first 300 hours.

With the flat plate configuration, five different waste streams were treated (1) lime clarified secondary effluent after dual media filtration and carbon adsorption, (2) lime clarified secondary effluent after dual media filtration, (3) lime clarified secondary effluent, (4) secondary effluent, and (5) primary effluent. Perhaps the most revealing result comes from comparison of the runs with Feeds 1 and 2 which had initial fluxes of 12 to 15 gal./ft^2•day. Feeds for both runs (1 and 2) were passed through a nominal 5-micron cartridge filter just before the module. Pertinent feed analyses are given in Table 2.

Table 2. Tests with Secondary Sewage Effluent in Plate and Frame Module

Run No.	Feed	TOC, mg/ℓ	Turbidity JTU	$b = -\frac{\Delta \log \text{flux}}{\Delta \log \text{time}}$
1	lime clarified, dual media filtration, carbon treated	1.0	0.5	0.05
2	lime clarified, dual media filtration	10.6	1.3	0.14

The greatly increased flux decline observed in the run with feed No. 2 (which had 10 times the total organic carbon of feed No. 1) was interpreted as supporting a positive role for soluble and colloidal organic carbon in causing flux decline.

In the test with feed No. 3 (lime-clarified secondary effluent) the value of the flux decline parameter, b, ($b = -\Delta \log \text{flux}/\Delta \log \text{time}$) was maintained at 0.03 by flushing every other day for one hour with an enzyme-detergent solution. With feed No. 4 (secondary effluent) a one-hour flush every day maintained a value of b = 0.04. In both cases the initial flux was ~8 gal./ft^2•day. A solution with greater than 35 mg/ℓ enzyme and 3500 mg/ℓ detergent was required for an effective flush.

With feed No. 5 (primary effluent) a daily flush with enzyme + detergent was only able to maintain a value of b = 0.19 starting with an initial flux of ~7 gal./ft^2•day.

The significance of the flux decline parameter, b, (cited above and used in subsequent portions of the report) can be appreciated by calculating the average flux for a one-year period of operation assuming that the flux decline parameter is constant throughout the year. Results of such a calculation for a range of b-values are given in Table 3.

Table 3. Average Value of Flux for One Year of Operation Assuming a Constant Value of Flux Decline Parameter Throughout the Year

$b = -\frac{\Delta \log \text{flux}}{\Delta \log \text{time}}$	Average Flux as % of Initial Flux
0.3	24.4
0.2	38.3
0.1	61.7
0.06	74.7
0.04	82.3
0.02	90.7
0.01	95.2
0.005	97.6

Nusbaum, Sleigh and Kremen (8) used spiral wound modules exclusively with carbon-treated secondary effluent rechlorinated to give approximately 2 mg/ℓ chlorine residual. Using daily air and water flushes and weekly flushes with a solution of 10,000 mg/ℓ of an enzyme containing laundry presoak product, flux decline parameters of 0.047 to 0.061 were obtained in 1000-hour tests with modules having an initial flux of 13 gal./ft^2•day and flux decline parameters of 0.055 to 0.077 were obtained in 1000-hour tests with modules having an initial flux of 17 gal./ft^2•day when the COD of the feed was 3 to 9 mg/ℓ. When the COD increased to 9 to 15 mg/ℓ, the flux decline slope of the higher flux modules was 0.14, indicating a direct relation between the rate of membrane fouling and the increase in COD of the feed.

Other studies showed that neither smaller brine side spacer thickness nor vertical module operation significantly affected module performance.

Beckman et al. (9) made further studies with spiral wound modules. Feed was from an oxidation pond after secondary treatment. Particularly notable were a series of three 200-hour tests at 750, 600, and 400 psig resulting in initial fluxes of 26, 22, and 14 gal./ft^2•day. Apparently no flushing procedures were used in these tests. Severest flux decline was observed with the highest flux membrane; after 90 hours the production rate from this membrane crossed below the rate for the lowest initial flux membrane. The intermediate flux membrane crossed below the lowest flux membrane after ~35 hours of operation.

Another important result of this study (9) was the discovery that sodium perborate and ethylenediaminetetraacetic acid as well as enzyme detergent flushes restored the flux to 80 to 85% of the initial value.

Recently, high flux cellulose acetate membranes have been developed (10). Three different formulations were developed: (1) a cellulose diacetate of moderately-low acetyl content (2) a cellulose triacetate-diacetate blend and (3) a crosslinked cellulose acetate methacrylate. All produced fluxes greater than 60 gal./ft^2•day and rejected at least 60% of a sodium chloride and 95% of a sodium sulfate solution when tested at 600 psig with 1000 mg/ℓ feed solutions. The performance of the membranes was evaluated using secondary effluent containing 5 ml per gallon of 5% sodium hypochlorite; pH was adjusted to 5.0 ± 0.25 with dilute sulfuric acid. Rejections with this feed are given in Table 1.

Flux decline was measured in 5- to 17-day tests with a 3-in. flat-plate test cell constantly recirculating the feed from a 5-gallon reservoir. Starting from an initial flux of ~50 gal./ft^2•day, the flux declined to less than 20 gal./ft^2•day in ~100 hours. With these high flux membranes, flux declines using carbon-treated secondary effluent was modest after 30 hours (from 65 to 55 gal./ft^2•day) but after ~100 hours flux was only ~20 gal./ft^2•day. Periodic flushing with an enzyme detergent largely prevented the flux declines in 100-hour tests.

In a general discussion of design considerations for treatment of solids-laden wastewaters by reverse osmosis, Feuerstein and Bursztynsky (11) present an interesting comparison of the effect of feed water quality on flux decline under comparable, controlled test conditions. The only pretreatment given to several grades of municipal wastewater was pH adjustment to ~5.0. Flux decline slopes, determined by replotting a graphical comparison of flux declines, are given in Table 4. The membranes had initial fluxes of 15 to 30 gal./ft^2•day.

Table 4. Flux Decline Slopes Obtained under Identical Conditions with Different Types of Municipal Wastewater [after Feuerstein and Bursztynsky (11)]

Sewage Type	$b = -\frac{\Delta \log \text{flux}}{\Delta \log \text{time}}$
1. Carbon-treated secondary	0.14
2. Secondary	0.35
3. Primary	0.56
4. Raw	~0.9

A novel scheme has been developed for preventing fouling of tubular modules when treating raw sewage which had been filtered through a 100-mesh screen (12). Before introducing sewage into the reverse osmosis system

the membrane is coated with diatomaceous earth, powdered activated carbon and surface active agent. This precoat mixture is applied under pressure; after seven hours 10 minutes of operation the pressure is dropped, and the accumulated solids are flushed from the system for 15 minutes. Then a cleaning solution of ammonium citrate and enzyme detergent at pH 5.5 is recycled for 15 minutes, the membrane is recoated and raw sewage introduced to begin the cycle again. No information was given about membrane lifetimes achievable.

Results to date have shown that considerable pretreatment beyond the secondary stage is required to prevent severe flux decline in treatment of municipal sewage effluent by hyperfiltration using commercial modules at flow rates recommended for brackish waters. Even with carbon treatment after the secondary stage, the rate of flux decline of membranes with different initial fluxes, increases as initial flux increases. The evidence is that soluble or colloidal organics are an important factor in flux decline with these highly treated sewage effluents.

Relatively long term runs have been achieved with carbon treated secondary effluent as feed by using frequent flushes with solutions of enzyme detergent, sodium perborate or ethylenediaminetetraacetic acid.

Extensive studies of cellulose acetate membranes operating on untreated river water feeds containing 10-100 mg/ℓ suspended solids have shown that flux decline can be arrested by operation at high tangential velocities (13). For example, after an initial flux decline during the first day's operation (from 36 to 26 gpd/ft^2), flux decline was arrested for the next nine days by the high axial velocity (24 ft/sec) used and after the first 10 days operation, the flux was still greater than 23 gpd/ft^2. When the axial velocity was dropped to 1.6 ft/sec, there was catastrophic flux decline during the next two days from 23 to 4 gpd/ft^2. Returning the velocity to 24 ft/sec for one day gave no significant flux recovery.

With some feeds velocities of 24 ft/sec may not be required to arrest flux decline. In one experiment velocity was reduced from 24 to 9.8 ft/sec after two days operation and from 9.8 to 4.9 ft/sec after six days operation. After two days operation at 4.9 ft/sec (eight days after the start of the run) flux was still greater than 23 gpd/ft^2 and there were no marked changes in the rate of flux decline as the velocity was successively changed. When the velocity was decreased to 1.6 ft/sec, there was a rather abrupt decline in flux from 23 to 13 gpd/ft^2 followed by a slower decline to 11 gpd/ft^2 during the next three days.

The results of flux decline studies frequently produce straight lines when plotted as log flux, J(t), vs log time, t (at constant pressure and temperature) i.e.,

$$J(t) = J_I \left(\frac{t}{T_I}\right)^{-b} , \qquad (1)$$

where J_I = reference flux at reference time, T_I. Thus, small values of the exponent "b" correspond to small values of flux decline with time.

These studies showed that mean values of b (when using Tennessee River water as feed) increased as the axial velocity decreased. At a velocity of 24 ft/sec, values of b were small (0.02 to 0.03) and were consistently in a narrow range regardless of feed composition. However, at smaller velocities, values of b were not only much larger (a mean of 0.09 to 1.6 ft/sec) but the range was much larger (from 0.07 to 1.0 at 1.6 ft/sec). In view of the poor reproducibility of results at the lower velocities, it appears that in this range flux is very sensitive to composition of the feed. At sufficiently high velocities, however, it seems that flux decline can be held to low values and is insensitive to feed composition for a particular wastewater stream.

Because of the indications that proper hydrodynamic conditions could markedly reduce the rate of flux decline with polluted surface water feeds, a program was initiated at the Oak Ridge National Laboratory to determine the effectiveness of various hydrodynamic and related techniques for control of fouling and flux decline of cellulose acetate membranes to improve the proposed hyperfiltration (reverse osmosis) method for treatment of municipal sewage effluents. Effluents from the primary and secondary stages of the Oak Ridge Sewage Disposal Plant were to be used. Since this sewage is of unusually low salinity, it was to be spiked with salts to facilitate analysis for salt rejection. Since it was anticipated that, under proper hydrodynamic conditions, it would be no more difficult to handle primary sewage effluents than secondary sewage effluents, emphasis in this program was to be on primary and treated primary effluents.

It was anticipated that, in the absence of turbulence promoters, high tangential velocities would be required to minimize flux decline and fouling. It was proposed to evaluate the effectiveness of this high tangential velocity technique using commercial cellulose acetate membranes of various intrinsic fluxes in test sections designed locally to allow good hydrodynamic control and ease of assembly. In one of these test sections the membrane is mounted on the outside of a cylindrical porous tube and this "finger" is inserted into a transparent pressure jacket which allows visual observation of the membrane during operation.

The principal parameters to be studied in the high tangential velocity systems were circulation velocity, time, and the effect of permeation rate on flux decline or fouling rate at velocities ranging from 5 to 20 ft/sec.

SECTION IV

EQUIPMENT

Mobile Hyperfiltration Loop. All tests were carried out in a test loop located at the Oak Ridge East Treatment Plant. The loop was substantially the same as that developed and used for an earlier FWQA program on application of dynamic membranes to treatment of municipal sewage effluents by hyperfiltration (5). The loop is mounted in a highway trailer having internal dimensions of 6 1/2 x 7 1/2 x 28 1/2 ft. Feed is circulated by a canned rotor, oil-cooled pump rated at 120 gpm with 294 ft head for fluid at 68°F. Pressurization and feed makeup are provided by a Mil-Roy Triplex pump rated at 4 gpm at 2000 psig. Pressure is controlled by an air-controlled 1-in. letdown valve. Volume of the loop and two test sections is about 5 gallons.

The test loop has a concentric pipe heat-exchanger to remove pump heat, two 1-in. test sections, and a test section by-pass to permit operation of the pump at near rated output even with low flows through the test sections. Flows through the test sections are controlled by ball valves located in each section and are measured by Venturis which have a range of 6 to 60 gpm.

Auxiliary equipment consists of a cooling oil system for the circulating pump, product tank, feed pump supply tanks, transfer pump between product and feed tanks, and an air compressor. Chemical addition to the loop is through either an air operated piston pump, used for intermittent additions, or a small diaphragm pump, used for continuous addition and supplied by a 50-liter tank. A skid mounted feed booster pump transfers primary effluent from downstream of the primary settling tanks to the feed pump supply tanks. Secondary effluent, supplied through a second skid-mounted pump, is used for the heat exchanger cooling water. Fresh water can also be supplied to the heat exchanger through a tank connected to the coolant pump suction line.

Test Sections. Two types of test sections were used. One similar to that shown in Figure 1 consists of a 5/8-in. test finger that has two 6-in. long porous sections fabricated from sintered stainless steel frit enclosed in a transparent jacket. The product sides of the test section are separated and have their own discharge lines. A cellulose acetate membrane is wrapped around each porous section and is sealed in place with pressure-sensitive, water-proof tape. The feed stream flows through the annulus between the test section and the jacket. The product stream flows through the membrane and is discharged at atmospheric pressure to the product tank.

The other type of test section is of similar design except it contains three porous sections each of which is sealed from the others. Three product lines are required. The pressure jacket is fabricated from 304L stainless steel. Since these pressure jackets are longer than the ones used for the two-section fingers, stainless steel was used as a safety measure to protect against failure in case of an accidental pressure surge in the loop.

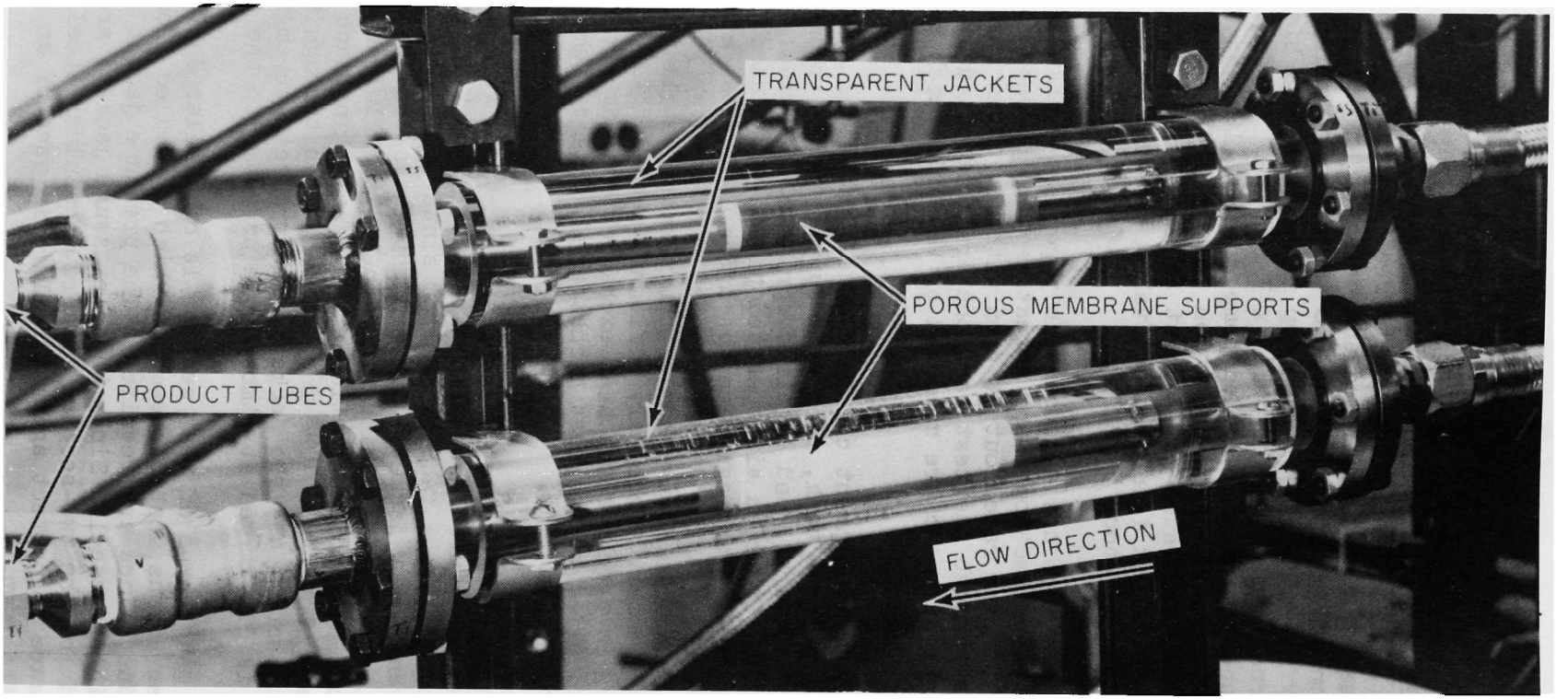

Figure 1. Hyperfiltration Test Section with Transparent Pressure Jacket.

SECTION V

FEED COMPOSITION AND CHEMICAL ANALYSIS

The composition of primary sewage effluent from a municipal sewage treatment plant varies during the day and also appears to have some seasonal trends. The Cl^- ion concentration and the turbidity (JTU) were monitored almost daily when the loop was operating by sampling the feed storage tank. A more complete analysis was made at less frequent periods. Figure 2 shows the data obtained during the period July 1, 1970 to August 1, 1971. Run numbers for tests in the present program are shown at the top of the figure with lines indicating the dates the runs were started and terminated. The temperature data are for the circulating fluid in the loop but reflect changes in sewage temperature since secondary effluent is used as the loop coolant.

No storm sewers are connected to the treatment plant. However, ground water does leak into the lines so that rains do affect the quantity and composition of primary effluent.

During the 13-month period that the primary sewage effluent was monitored, the turbidity varied between 5 and 66 JTU with slightly higher values during warm weather and following hard rains. Light rain tended to reduce turbidity. Inorganic carbon concentration was relatively constant varying between 32 and 60 mg/ℓ, with lower values occurring during cold weather. The amount of organic carbon varied considerably more ranging from 15 to 118 mg/ℓ. Phosphate concentration varied from 5 to 67 mg/ℓ. Normally, samples were obtained in mid-morning when phosphate concentrations were 20 to 50 mg/ℓ so that rejection values for membranes would be more accurate. Ca^{++} and Mg^{++} concentrations remained fairly constant — 0.00112 to 0.0017 meq/l — showing little seasonal trend. The Cl^- ion concentration was always less than 1.0 meq/l.

Chloride analysis was performed amperometrically with a Buchler-Cotlove chloridometer. Turbidity was determined with a Hach 2100 turbidimeter. Ca^{++} plus Mg^{++} concentrations were measured together by EDTA titration using Eriochrome Black T indicator and potassium cyanide as an inhibitor for metal ions. Total phosphate was determined by a colorimetric method using aminonapthosulfonic acid during the first part of the program. Then stannous chloride was used as the reducing agent during the latter part of the program because of its greater sensitivity. Total and inorganic carbon were determined with a Beckman 915 analyzer. Organic carbon was obtained as the difference between these determinations.

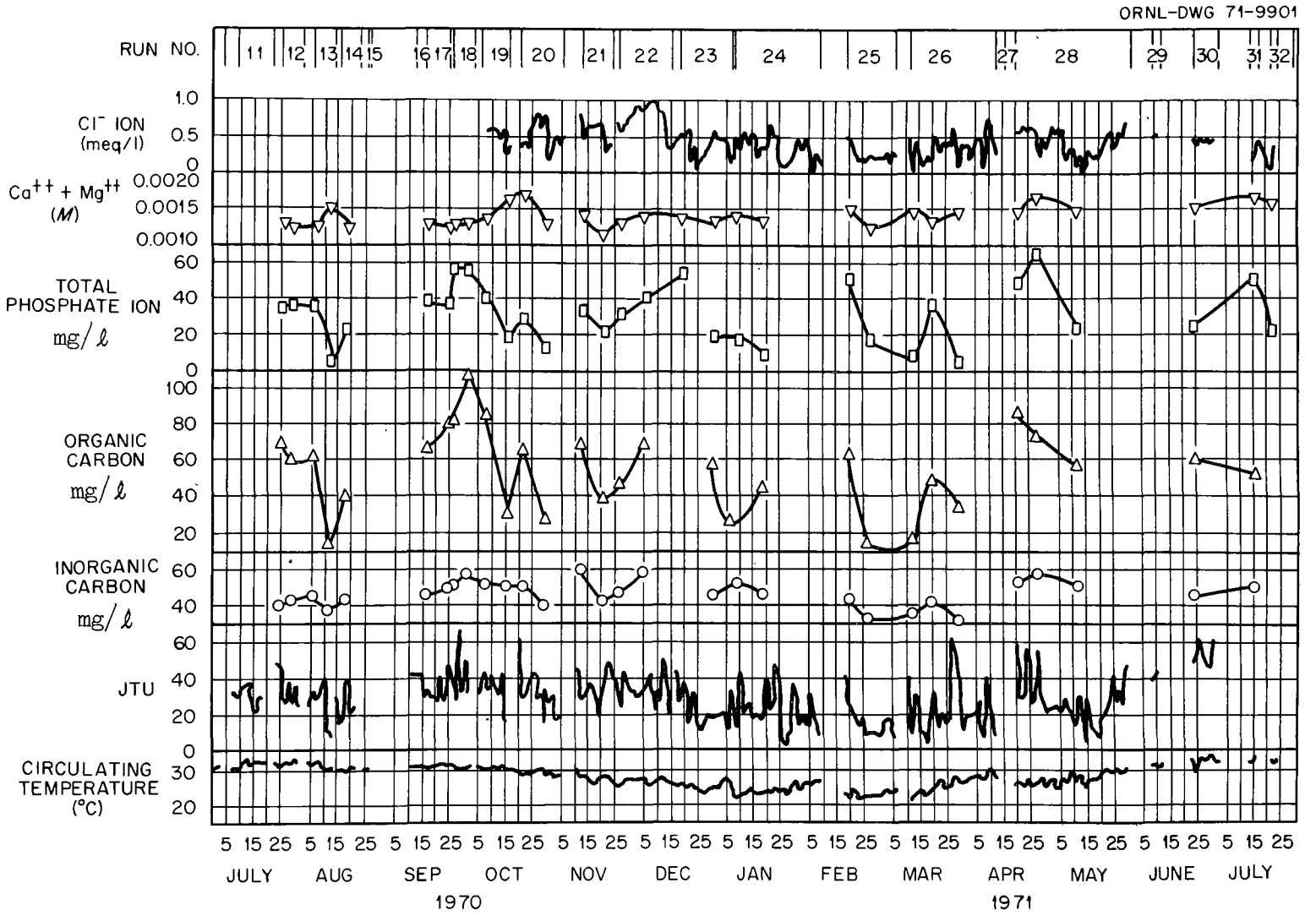

Figure 2. Variation of Constituents in Primary Sewage Effluent, Oak Ridge East Sewage Plant.

SECTION VI

RESULTS

Three types of membrane with markedly different characteristics were used in this study: (1) an unheat-treated membrane (Eastman HT-00) with a nominal initial flux of 150 to 200 gal./ft^2•day at 600 psig and nominal zero rejection, (2) a membrane (Aerojet 6%) with nominal initial flux of 40 to 50 gal./ft^2•day at 600 psig and chloride rejection of ~94%, and (3) a membrane (Eastman) heat-treated at ~90°C to give a nominal initial flux of 2 to 10 gal./ft^2•day and 97 to 99% chloride rejection. With our experimental equipment, three sections of a membrane were always exposed to the same feed at three different velocities. In the first 10 tests, two of the three membrane types were compared simultaneously; in the remainder of the tests all three membrane types were compared simultaneously at three different velocities, i.e., three membranes each exposed to three different velocities or 9 tests per run.

Before starting a run the test loop was cleaned by circulating two batches of 1 M NaOH for one hour each; then the loop was rinsed with city water until the pH of the rinse water was 7. At the start of every run, membranes were exposed for 2 to 18 hours to a feed of city water spiked with NaCl to obtain baseline performance for the membrane. The flux one-half hour after startup on city water was taken as the initial flux, J_1.

The runs made during this study are summarized in Tables 5 and 6. Table 5 gives the total run time in hours, the velocity at which the membranes were exposed, the duration of the baseline city water portion of the test and the flux and observed rejection for chloride measured one-half hour after startup with city water. The bulk of the table gives flux and chloride rejection at 20, 50, 100, 150, and 200 hours after switching to primary sewage feed. Samples were taken at 20 and 200 hours for analysis for PO_4^-, Ca^{++} plus Mg^{++}, organic carbon, and inorganic carbon. These latter results will be summarized in a subsequent part of the report. Since unscheduled shutdowns frequently may affect flux and rejection, the time of occurrence and duration of the shutdowns is given in the last two columns of Table 5. A few tests were extended to times longer than 200 hours, results for the extended time portion of the tests are given in Table 6.

Flux decline results are frequently presented as log flux vs log time or as log flux vs arithmetic time. The first form implies the possibility of a power law dependence according to Equation (1) while the second implies the possibility of an exponential dependence:

$$\frac{J(t)}{J_o} = e^{-Bt} \quad . \qquad (2)$$

Since there is no theory for fouling which supports either Equation (1) or Equation (2), it is immaterial which form is used in presenting experimental results. More important, great caution should be used in extrapolating results for flux decline for any appreciable time beyond that for which data are available using either Equation (1) or Equation (2). As will be illustrated with the present results, apparent linear relations may be followed on one or the other of the two types of plot for hundreds of

Table 5 . Summary of Cellulose Acetate Runs with City Water and Primary Sewage Feed[17]

Run No. (Starting Date)	Total Run Time, Hr	Memb.[16]	Vel. fps	Water Run: Run Time, Hr.	Water Run: 1/2 Hr. gfd	Water Run: 1/2 Hr. R_{obs} %	Time After Change to Primary Sewage: 20 Hr. gfd	20 Hr. R_{obs} %	50 Hr. gfd	50 Hr. R_{obs} %	100 Hr. gfd	100 Hr. R_{obs} %	150 Hr. gfd	150 Hr. R_{obs} %	200 Hr. gfd	200 Hr. R_{obs} %	Shutdowns: Total Run Time, Hr.	Shutdowns: Duration Hr.
13 (8-4-70)	177	B	15.3	2	56.1	82.6	40.0	94.6	38.7	93.6	34.0	94.0	32.2	95.3	31.4[1]	92.0[1]	69 2/3	1 3/4
			9.9		74.3	66.2	34.2	93.1	33.9	93.2	29.9	93.1	23.1	93.8	10.7	91.8	129 3/4	13 1/2
			5.2		68.5	72.8	11.2	83.8	6.5	79.3	4.1	70.0	10.0	87.1	5.2	82.0		
		A	15.3		199.8	11.2	91.6	27.8	83.5	23.2	77.5	19.4	66.6	22.5	57.5	24.2		
			9.9		171.3	8.7	35.7	34.0	29.0	29.0	25.2	25.8	17.5	28.5	12.1	31.6		
			5.2		180.6	0	16.3	16.0	11.3	16.0	8.7	16.0	4.7	19.0	3.3	18.0		
14 (8-14-70)	156	B	20.0	2	52.2	86.3	31.5	94.6	29.9	94.7	31.6	94.8	30.6	95.5			35 1/2	10 2/3
			13.6		56.0	80.0	33.7	93.6	30.9	93.6	31.0	93.6	31.0	95.4				
			7.2		61.9	68.4	27.0	92.0	15.4	92.0	12.7	92.0	12.7	91.3				
		A	20.0		495.5	2.0	40.3	11.1	20.3	10.8	13.9	18.2	11.1	24.7				
			13.6		180.7	12.5	74.0	23.0	58.0	25.2	51.4	31.4	38.6	35.8				
			7.2		166.5	18.2	33.0	29.1	19.2	34.0	12.1	32.0	9.4	30.8				
17[2,3] (9-17-70)	190	B	30.0	6 1/2	61.9[4]	75.3[4]	31.9	93.5	29.7	93.6	26.9	93.8	26.1	94.8	24.5[1]	95.4[1]	27	
			7.6		336.1	16.2	16.9	92.0	10.7	90.4	7.5	89.0	10.7	91.3	8.6	91.0		
			4.0		213.1	24.2	9.6	82.0	5.1	78.8	3.3	75.9	2.9	71.8	2.4	70.3		
		A	30.0		211.1	10.8	89.4	22.1	84.8	25.8	82.0	28.7	71.5	41.5	71.3	44.0		
			7.6		434.9	7.0	13.7	44.2	9.2	44.2	6.8	44.2	5.5	44.2	4.4	44.5		
			4.0		220.2	9.6	9.5	40.1	5.2	40.0	3.3	39.2	2.6	39.0	1.9	37.7		
18 (9-25-70)	167	B	30.0	1 3/4	53.4[5]	89.9[5]	36.2	94.5	33.9	94.6	32.0	94.7	31.0	91.8[6]	30.3[1]	91.8[1]	144 1/2	1 min.
			12.0		49.9	87.8	33.1	94.8	27.0	94.7	24.7	94.6	22.8	94.5	22.0	96.3		
			6.4		60.0	76.2	18.7	92.2	10.4	90.4	6.7	88.6	5.2	87.4	4.0	87.7		
		A	30.0		151.3	21.6	100.0	36.6	82.1	39.8	71.2	43.0	65.6	44.0	60.2	51.8		
			12.0		138.2	0	10.8	15.9	10.2	18.0	9.6	20.1	9.3	21.4	9.2	24.0		
			6.4		158.7	6.9	22.3	34.0	11.3	34.0	8.2	34.0	6.2	34.0	5.2	28.5		
19 (10-5-70)	235	B	15.3	19[7]	37.2	97.1	23.7	97.9	22.8	97.9	22.2	98.0	21.9	98.0	21.6	98.0		
			9.9		39.5	92.4	24.5	97.9	23.9	97.9	23.0	98.0	22.7	98.0	22.3	98.0		
			5.2		49.4	77.4	14.1	97.8	9.1	97.5	6.5	97.3	5.4	97.2	4.7	97.1		
		A	15.3		137.6	1.0	10.8	8.0	9.7	10.5	9.0	12.0	8.6	14.0	8.4	15.5		
			9.9		80.3	44.9	42.1	46.5	37.9	35.7	30.9	36.0	27.7	36.3	25.4	37.0		
			5.2		87.1	31.0	11.3	72.0	6.3	74.0	4.1	58.0	3.2	56.0	2.6	54.5		

Table 5 . (Continued)

Run No. (Starting Date)	Total Run Time, Hr	Memb.[1e]	Vel. fps	Water Run: Run Time, Hr.	Water Run: 1/2 Hr. gfd	Water Run: 1/2 Hr. R_{obs} %	Time After Change to Primary Sewage: 20 Hr. gfd	20 Hr. R_{obs} %	50 Hr. gfd	50 Hr. R_{obs} %	100 Hr. gfd	100 Hr. R_{obs} %	150 Hr. gfd	150 Hr. R_{obs} %	200 Hr. gfd	200 Hr. R_{obs} %	Shutdowns: Total Run Time, Hr.	Shutdowns: Duration Hr.
20[e] (10-19-70)	342	B	12.0	18	50.6	92.7	36.8	95.2	36.0	95.5	35.3	95.8	33.3	95.9	32.1	96.0	244	14 1/3
			9.0		60.6	77.0	34.1	93.3	33.3	94.6	32.3	95.4	29.0	95.8	27.7	94.2	304 1/4	11 1/2
			4.75		50.7	89.0	24.3	85.0	10.0	83.1	5.1	75.5	3.4	69.4	2.6	64.0	315 3/4	11 1/2
		A	12.0		163.3	2.0	35.0	6.0	38.0	10.0	40.2	10.0	38.8	9.0	39.3	7.6		
			9.0		181.7	7.0	53.0	20.0	47.4	20.5	38.8	28.4	27.4	24.7	21.7	24.8		
			4.75		184.4	4.0	15.4	25.5	7.5	29.0	4.4	31.8	3.2	27.9	2.5	27.0		
21 (11-11-70)	288	C	15.3	3 1/2	2.1	22.0	0.8	90.0	0.8	92.5	0.7	94.1	0.7	94.8	0.7	95.3		
			9.9		0.7	38.5	0.7	90.8	0.7	92.7	0.7	93.7	0.6	94.3	0.6	94.7		
			5.2		0.7	48.0	0.6	97.0	0.6	98.5	0.6	99.1	0.6	99.3	0.6	99.0		
		A	15.3		129.4	27.0	71.5	43.0	66.5	44.0	62.3	44.1	61.7	44.2	60.0	44.3		
			9.9		141.0	20.3	40.5	42.0	37.8	42.2	35.7	42.4	34.8	42.5	34.0	42.6		
			5.2		125.2	21.5	13.0	44.5	6.5	39.5	4.0	36.3	2.9	34.5	2.3	33.0		
22 (11-25-70)	444	C	12.0	2 1/2	7.6	76.0	4.3	97.5	4.3	99.8	4.1	99.9	4.3	99.5	4.2	99.6	131	13
			5.7		16.6	26.6	3.4	70.0	3.3	70.0	3.2	70.0	3.7	56.0	3.2	52.0		
			3.0		20.4	16.7	3.0	91.6	2.9	94.1	2.8	95.6	2.8	95.7	2.5	97.9		
		A	12.0		156.6	21.4	51.7	50.8	47.5	51.0	44.8	51.1	41.8	53.0	33.4	57.0		
			5.7		133.0	1.0	9.6	26.5	4.8	25.2	2.9	24.0	2.4	23.5	1.9	23.0		
			3.0		126.4	5.4	4.9	30.0	2.9	28.5	1.9	28.0	1.8	27.5	1.5	27.0		
23 (12-17-70)	479	C	12.0	3	30.0	18.8	5.0	93.4	4.7	95.4	4.5	96.5	4.4	97.0	4.4	97.3		
			5.7		13.6	17.0	3.4	96.0	3.3	97.4	3.3	98.1	3.1	97.8	2.8	97.5		
			3.0		11.6	41.3	4.1	95.4	3.6	96.4	3.2	97.0	2.8	97.3	2.2	95.6		
		A	12.0		122.4	31.0	40.0	61.2	38.0	60.4	36.7	29.5	34.1	28.7	30.0	28.0		
			5.7		115.6	13.0	11.6	41.6	6.2	36.7	3.3	33.2	2.3	31.0	1.8	29.5		
			3.0		123.2	5.6	5.7	31.9	3.6	29.5	2.2	27.0	1.8	26.9	1.5	24.5		
24[g] (1-7-71)	752	B	15.0	2 1/2	41.7	92.9	34.2	56.0	32.6	57.2	31.5	58.1	30.8	59.0	30.0	59.4	299	14
			9.9		47.4	78.9	35.5	38.0	33.0	42.5	31.2	46.0	30.1	47.5	29.5	48.5	631	3/4
			5.2		40.4	91.5	6.4	86.3	4.3	82.5	2.9	78.7	2.3	75.5	1.9	70.0		
		A	15.0		146.5	18.7	80.4	39.5	77.2	39.5	75.6	39.5	69.7	39.5	65.0	39.5		
			9.9		141.9	10.3	38.5	38.0	36.0	36.1	34.3	35.0	30.2	34.2	27.7	34.0		
			5.2		122.0	9.8	8.30	44.5	3.8	44.0	2.6	43.5	2.1	43.3	1.8	43.0		

Table 5 . (Continued)

Run No. (Starting Date)	Total Run Time, Hr	Memb.[1e]	Vel. fps	Water Run: Run Time Hr.	Water Run: 1/2 Hr. gfd	Water Run: 1/2 Hr. R_{obs} %	Time After Change to Primary Sewage: 20 Hr. gfd	20 Hr. R_{obs} %	50 Hr. gfd	50 Hr. R_{obs} %	100 Hr. gfd	100 Hr. R_{obs} %	150 Hr. gfd	150 Hr. R_{obs} %	200 Hr. gfd	200 Hr. R_{obs} %	Shutdowns: Total Run Time, Hr.	Shutdowns: Duration Hr.
25 (2-18-71)	352	C	17.6	2 1/2	4.4	80.5	2.9	99.3	2.8	99.9	2.7	99.8	2.7	99.7	2.8	99.6	303 1/3	7 1/2
			10.0		5.7	60.0	2.9	99.0	2.9	99.7	2.8	99.6	2.8	99.5	2.9	99.4		
			3.1		11.7	77.4	3.1	98.7	2.7	99.1	2.5	98.7	2.6	98.5	2.4	98.2		
		B	17.6		81.3	48.6	36.0	95.6	34.2	96.5	30.7	96.3	29.1	95.8	28.0	95.4		
			10.0		44.6	84.5	35.0	95.3	33.5	95.6	29.4	95.7	26.5	95.2	24.8	94.9		
			3.1		43.0	77.6	4.4	67.2	2.5	65.8	1.8	64.5	1.6	63.6	1.5	62.9		
		A	17.6		81.7	52.5	64.0	72.1	60.6	74.2	55.5	75.7	51.8	76.6	49.3	77.1		
			10.0		117.2	25.9	45.0	50.2	42.3	50.4	30.2	49.7	25.9	49.4	19.0	49.2		
			3.1		101.0	21.5	4.3	41.0	2.6	40.2	1.8	40.1	1.6	40.0	1.4	40.0		
26 (3-11-71)	768	C	17.6	2 1/2	5.4	81.5	4.8	98.1	4.6	98.4	4.6	98.7	4.5	98.8	4.5	99.0		
			10.0		3.6	88.0	3.3	98.0	3.2	99.3	3.1	99.7	3.1	99.8	3.1	99.9		
			3.1		6.3	70.5	3.4	97.6	3.2	98.1	3.1	96.4	2.8	98.3	2.5	98.2		
		B	17.6		55.9	73.9	34.8	94.4	31.8	94.7	29.0	94.9	27.2	95.0	26.0	95.5		
			10.0		49.3	88.9	5.0	94.4	33.0	94.7	29.8	95.0	25.9	95.1	2.2	95.2		
			3.1		39.2	90.4	93.6	70.5	3.1	67.5	2.2	66.2	1.7	63.0	1.5	60.5		
		A	17.6		143.9	27.0	50.8	45.8	87.3	50.0	78.0	53.0	72.0	56.0	69.3	61.2		
			10.0		115.9	23.9	5.9	40.0	48.5	41.0	34.8	42.0	26.8	42.3	22.5	45.0		
			3.1		113.7	19.1		36.0	5.5	36.0	2.6	36.0	2.2	36.0	1.9	36.2		
28[3] (4-19-71)	985	C	17.6	2 1/4	2.8	92.6	2.4	99.5	2.4	99.9	2.4	99.8	2.3	99.8	2.4	99.9	208 5/6	7 1/2
			10.0		8.1	33.9	3.0	96.7	2.9	98.5	2.8	99.2	2.7	99.3	2.7	99.2		
			3.1		10.8	49.5	3.2	95.3	3.2	99.4	3.0	99.5	2.8	99.5	2.7	99.5		
		B	17.6		40.7	93.0	31.2	95.5	30.8	95.4	28.3	95.4	27.6	95.6	26.7	96.0		
			10.0		48.6	83.2	31.6	94.8	30.2	94.8	29.9	94.8	29.0	95.2	28.3	95.9		
			3.1		59.3	69.8	5.4	83.3	2.8	74.0	2.0	71.0	1.6	68.0	1.5	68.5		
		A	17.6		177.7	16.5	96.5	30.9	89.0	32.2	75.0	40.0	60.0	46.0	59.2	49.2		
			10.0		197.1	6.3	39.0	29.1	39.9	27.0	39.9	28.2	37.9	26.5	35.0	29.0		
			3.1		154.8	12.1	6.2	37.0	3.5	35.7	2.2	36.5	1.8	36.4	1.7	36.4		
29[10] (6-8-71)	72	C	17.6	2 1/4	4.4	51.0	3.1	99.2	3.1	99.5								
			10.0		3.6	92.6	2.9	98.2	2.7	98.3								
			3.1		15.3	22.8	3.2	98.2	3.4	99.2								

Table 5. (Continued)

Run No. (Starting Date)	Total Run Time, Hr	Memb.[16]	Vel. fps	Water Run: Run Time, Hr.	Water Run: 1/2 Hr. gfd	Water Run: 1/2 Hr. R_{obs} %	Time After Change to Primary Sewage: 20 Hr. gfd	20 Hr. R_{obs} %	50 Hr. gfd	50 Hr. R_{obs} %	100 Hr. gfd	100 Hr. R_{obs} %	150 Hr. gfd	150 Hr. R_{obs} %	200 Hr. gfd	200 Hr. R_{obs} %	Shutdowns: Total Run Time, Hr.	Shutdowns: Duration Hr.
29 (Continued)		B	17.6		75.8	67.9	38.0	94.7	36.8	94.0								
			10.0		56.6	92.1	26.7	91.9	26.9	91.6								
			3.1		76.8	60.9	10.9	78.2	4.4	73.7								
		A	17.6		192.6	16.7	71.5	29.0	70.0	27.8								
			10.0		204.3	10.7	28.8	24.5	24.7	28.3								
			3.1		167.5	15.9	11.3	30.0	4.3	31.0								
30[11] (6-23-71)	192	C	17.6	2 1/6	4.1	61.9	2.1	97.4	2.1	99.0	2.1	99.5	2.1	99.5	2.1[1]	96.0[1]		
			10.0		6.6	22.7	1.3	93.1	1.3	96.6	1.4	96.3	1.4	96.6	1.4	96.2		
			3.1		9.1	23.4	1.3	91.4	1.3	95.9	1.4	97.0	1.3	98.0	1.3	97.0		
		B	17.6		63.0	78.5	28.8	97.8	26.8	95.3	25.9	98.2	23.7	98.3	21.3	98.8		
			10.0		53.8	87.0	28.7	96.5	26.5	97.8	17.8	96.5	11.5	95.4	8.4	94.3		
			3.1		86.6	56.9	6.8	69.2	3.9	55.6	2.8	51.9	2.1	48.1	1.7	47.3		
		A	17.6		70.2	39.3	35.6	71.8	30.8	82.7	25.8	85.1	21.0	86.9	18.0	88.7		
			10.0		68.9	34.6	33.0	77.2	27.7	77.5	19.4	77.1	12.8	70.0	9.5	63.8		
			3.1		89.9	21.7	7.1	29.8	4.9	26.0	2.8	24.3	2.0	22.8	1.6	22.1		
31[13] (7-13-71)	58	C	24.0	11 1/2	13.1	12.4	0.5	87.0	0.3[1]	84.0[1]							10	7 1/2
			12.0		5.8	21.6	1.3	94.6	1.4	95.5								
			7.4		44.9	3.3	3.8	56.0	3.6	46.5								
		B	24.0		53.0	93.1	1.1	48.5	0.7	42.2								
			12.0		113.0	43.3	4.5	55.5	3.2	53.4								
			7.4		180.7	26.6	5.8	48.0	4.5	40.0								
		A	24.0		63.5	42.5	0.9	46.1	0.5	41.0								
			12.0		89.0	22.2	5.3	26.2	2.4	30.8								
			7.4		84.7	25.4	4.6	23.4	3.9	18.4								
32[14] (7-21-71)	51	C	19.1	3 1/2	3.9	68.3	2.2	91.9	2.2[1]	90.0[1]								
			6.7		2.6	95.9	2.0	82.0	1.9	87.0								
			2.9		3.6	69.2	1.8	90.9	1.8	85.0								
		B	19.1		50.5	93.5	33.0	92.9	23.7	94.7								
			6.7		55.2	89.8	22.5	88.6	14.2	93.6								
			2.9		57.2	87.8	8.6	82.5	4.4	74.0								

Table 5 . (Continued)

Run No. (Starting Date)	Total Run Time, Hr	Memb.[16]	Vel. fps	Water Run: Run Time, Hr.	Water Run: 1/2 Hr. gfd	Water Run: 1/2 Hr. R_{obs} %	Time After Change to Primary Sewage: 20 Hr. gfd	20 Hr. R_{obs} %	50 Hr. gfd	50 Hr. R_{obs} %	100 Hr. gfd	100 Hr. R_{obs} %	150 Hr. gfd	150 Hr. R_{obs} %	200 Hr. gfd	200 Hr. R_{obs} %	Shutdowns: Total Run Time, Hr.	Shutdowns: Duration Hr.
32[14] (Continued)		A	19.1		73.7	43.5	48.1	49.8	27.5	65.0								
			6.7		77.4	32.3	30.3	48.6	26.3	58.0								
			2.9		80.8	34.1	8.7	44.5	4.7	59.0								
33 (7-29071)	409	C	21.8	2	2.1	99.5	1.7	97.8	1.7	99.3	1.6	99.9	1.6	99.9	1.6	99.9	86	9 1/3
			8.0		2.5	91.6	2.2	98.4	2.2	99.2	2.2	97.8	2.2	97.9	2.1	99.0	159 1/2	1 min.
			3.3		2.0	92.8	1.5	92.4	1.7	86.8	1.6	93.8	1.4	98.5	1.5	98.9	288	14 1/6
		B	21.8		49.8	94.4	35.8	95.7	32.0	96.1	30.8	96.3	29.2	96.3	28.2	96.8		
			8.0		51.2	93.4	29.1	94.5	25.7	94.2	21.0	93.9	14.4	93.0	7.4	95.2		
			3.3		47.7	92.6	8.5	85.2	4.7	80.0	7.8	87.0	3.6	80.3	3.8	73.0		
		A	21.8		65.5	51.5	46.9	63.2	41.7	68.5	40.7	70.9	38.2	71.8	37.6	74.8		
			8.0		64.8	45.9	31.4	66.5	22.9	67.2	21.0	67.4	13.9	70.0	11.0	70.8		
			3.3		61.4	50.7	9.0	58.8	5.3	54.0	7.8	62.9	3.8	57.7	2.6	57.0		
34[16] (8-23-71)	48	C	9.0	3 1/2	7.5	32.5	1.7	97.4	1.4[1]	98.4[1]								
			5.0		8.2	17.3	1.6	97.2	1.5	97.9								
			3.0		7.5	21.0	1.9	94.0	1.7	97.6								
		B	9.0		54.6	94.5	21.8	86.3	9.6	89.9								
			5.0		54.6	10.2	15.0	88.4	8.3	86.6								
			3.0		71.6	68.2	12.7	88.5	5.7	85.4								
		A	9.0		64.8	49.9	25.9	60.0	12.3	67.1								
			5.0		71.6	38.2	15.0	34.2	7.5	51.2								
			3.0		75.0	36.9	13.1	30.8	6.3	48.5								
35[16] (8-31-71)	25	C	9.0	2	1.9	99.9	1.4	88.5										
			6.0		3.3	99.3	1.7	58.6										
			3.7		9.5	23.4	1.8	67.0										
		B	9.0		52.6	99.1	23.8	93.7										
			6.0		68.6	66.4	19.0	85.7										
			3.7		56.6	98.6	16.1	79.8										
		A	9.0		76.6	30.0	25.2	51.9										
			6.0		79.3	36.0	20.3	45.3										
			3.7		92.6	19.6	16.2	3.5										

Footnotes for Table 5. Summary of Cellulose Acetate Runs with City Water and Primary Sewage Feed

1. Extrapolated data.

2. Run 15 terminated prematurely because system pressure controller failed.

3. Run 16 terminated prematurely because of defective membranes.

4. One-and-one-half hour sample.

5. One-hour sample.

6. Seal on test section loosened at 118 hours.

7. 101 mg/ℓ polyvinylmethyl ether (PVME) circulated during water run.

8. Membranes soaked for three days in PVME before being installed.

9. Membranes soaked 30 days in polyacrylic acid (100 mg/ℓ) while in test section before testing.

10. Ferric chloride added during sewage circulation (66 mg/ℓ Fe).

11. Cat-Floc added during sewage circulation (20 mg/ℓ).

12. Eastman membranes from a different batch of material than used in previous tests.

13. Alum [commercial $Al_2(SO_4)_3 \cdot nH_2O$] added during sewage circulation (50 mg/ℓ Al).

14. Aqua Nuchar A added during sewage circulation (100 mg/ℓ).

15. Ferri Floc [commercial $Fe_2(SO_4)_3 \cdot nH_2O$] added during sewage circulation (~50 mg/ℓ Fe).

16. Membrane type, A: Eastman HT-00, B: Aerojet 6%;
    C: Eastman heat-treated @ 90°C.

17. All runs were made at a pressure of 600 psig.

Table 6.
Summary of Results at 400 and 700 Hours for Long Term Runs with Cellulose Acetate and Primary Sewage Feed (First 200 Hours of Runs Summarized in Previous Table)

Run No.	Membrane	Velocity ft/sec	Time After Change to Primary Sewage 400 Hours gfd	400 Hours R, %	700 Hours gfd	700 Hours R, %
22	Eastman	12.0	3.89	99.9		
	Heat Treated 90°C	5.7	1.79	41.5		
		3.0	1.07	96.8		
	Eastman	12.0	19.7	66.5		
	Unheat-Treated	5.7	1.1	22.3		
		3.0	0.9	29.6		
23	Eastman	30.0	4.08	97.6		
	Heat Treated 90°C	13.6	2.17	97.1		
		11.6	1.20	92.7		
	Eastman	30.0	23.1	68.1		
	Unheat-Treated	13.6	1.32	30.0		
		11.6	1.05	25.5		
24	Aerojet	15.0	24.3	95.9	19.1	97.4
		9.9	20.6	95.0	18.8	96.1
		5.2	2.14	73.5	2.37	84.1
	Eastman	15.0	39.2	49.0	25.4	54.5
		9.9	8.9	36.6	21.1	29.5
		5.2	1.43	39.5	1.18	56.7
26	Eastman	17.6	4.60	99.8	4.59	99.7
	Heat Treated 90°C	10.0	3.29	99.9	3.37	99.9
		3.1	1.21	97.5	0.74	98.0
	Aerojet	17.6	20.9	96.7	17.4	96.5
		10.0	16.3	95.6	12.9	95.3
		3.1	1.22	56.4	0.72	56.0
	Eastman	17.6	45.0	73.5	33.0	64.0
	Unheat-Treated	10.0	12.7	50.8	7.65	56.8
		3.1	1.22	36.5	0.81	39.0
28	Eastman	17.6	2.40	99.9	2.46	99.9
	Heat Treated 90°C	10.0	2.80	99.8	2.88	99.8
		3.1	2.07	99.2	1.28	98.9
	Aerojet	17.6	24.5	96.1	21.7	96.7
		10.0	27.8	95.1	23.8	96.4
		3.1	1.03	65.3	0.89	61.0
	Eastman	10.0	29.9	32.1	21.2	40.0
	Unheat-Treated	3.1	1.12	34.0	1.00	34.0
33	Eastman	21.8	1.64	99.9		
	Heat Treated 90°C	8.0	2.21	99.8		
		3.3	1.42	99.4		
	Aerojet	21.8	23.9	96.6		
		8.0	5.11	88.9		
		3.3	1.42	73.0		
	Eastman	21.8	30.1	81.8		
	Unheat-Treated	8.0	5.11	64.4		
		3.3	1.50	55.0		

hours before marked deviations become apparent. For this reason we prefer as a definition of the flux decline parameter, b:

$$b = -\frac{\Delta \log \text{flux}}{\Delta \log \text{time}} \quad . \tag{3}$$

With this definition, b may be a function of time and there is no implication of what the functional relation between "b" and "time" might be.

The bulk of our results will be presented as composite plots of data from many runs. This approach was chosen because the inherent day-to-day and week-to-week variation of sewage composition introduces an unknown variability in the results. Although the number of tests made was insufficient to represent a good statistical sample, enough data were obtained to give an idea of the range of results which might be expected under different operating conditions.

Before presenting the composite results, several individual runs will be discussed because they illustrate a particular point or because special treatment was given to the membrane or to the primary effluent feed.

Reproducibility of Results

Run 26. Figure 3 shows typical results of a single run, using primary sewage as feed, in which three different membrane types were exposed simultaneously to three different axial velocities. This test is identified as Run 26 in Tables 5 and 6. The three membranes used were Aerojet-General 6%, Eastman HT-00 as received (unheat-treated), and the same type membrane heat-treated in water for five minutes at 90°C. Axial velocities in the test sections were 17.6, 10, and 3.1 ft/sec. Loop pressure was maintained at 600 psig. Temperature of the circulating fluid ranged between 21.2 and 28°C showing an increasing trend with time resulting in part from the increasing temperature of the cooling water to the heat exchanger. One hour after the start of sewage circulation a high pressure excursion shut the loop down. The cause of the excursion was not determined. The run was restarted in about one minute. The shutdown, which caused the system pressure to drop quickly to 0 psig, did not appear to affect the operating characteristics of the membrane at this initial period of circulation.

City water spiked with NaCl to ~0.01 M was circulated for the first 2 1/2 hours of the test to determine the initial flux and rejection for each membrane before the feed was changed to primary sewage effluent. Reference fluxes, J_I, and rejections are shown in Table 7.

Initial fluxes for the heat-treated Eastman membranes ranged from 3.6 to 6.3 gfd with rejections (71 to 88%) that varied inversely with flux. No velocity dependence is apparent. Flux for the Aerojet membranes ranged from 39 to 56 gfd increasing as the axial velocity increased. Rejections, which varied from 74 to 90%, decreased as the flux increased. For the unheat-treated Eastman membranes the flux (114-144 gfd) and the rejection (19-27%) increased as the axial velocity increased.

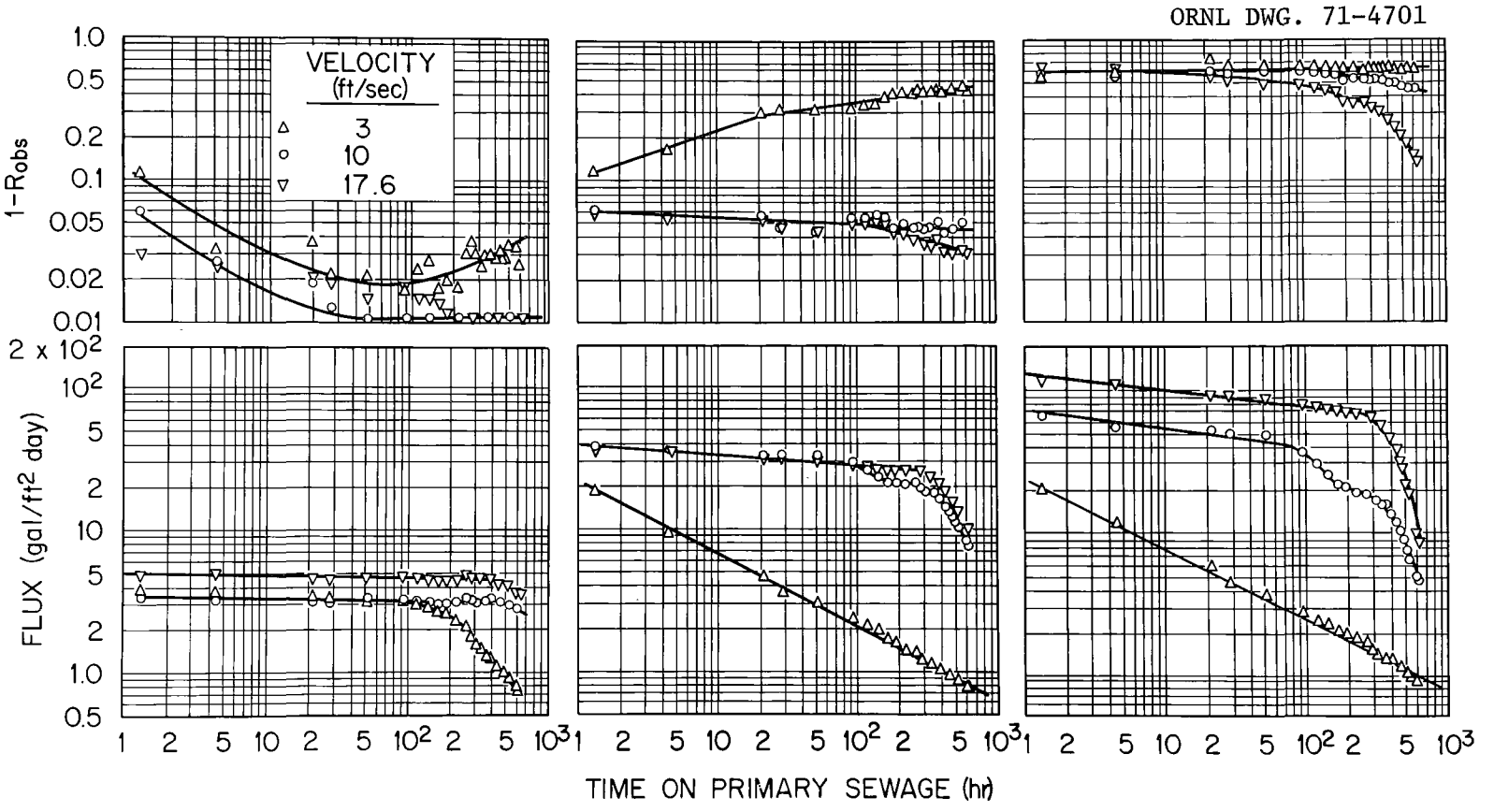

Figure 3. Effect of Flux and Axial Velocity on Flux Decline Using Primary Sewage as Feed. (Cellulose Acetate Membrane, 600 psig, Run 26)

Table 7. Reference Flux and Rejection (Run 26, System Pressure, 600 psig)

Membrane	Velocity ft/sec	J_I gfd	R_{obs} %
Eastman HT-00 Heat-treated 90°C	17.6	5.4	82
	10	3.6	88
	3.1	6.3	71
Aerojet-General 6%	17.6	56	74
	10	49	89
	3.1	39	90
Eastman HT-00	17.6	144	27
	10	116	24
	3.1	114	19

Flux and rejection as a function of axial velocity and time after the introduction of primary sewage effluent are shown in the left portion of Figure 3 for the heat-treated Eastman membranes, in the middle portion of Figure 3 for the Aerojet membranes, and in the right portion of Figure 3 for the unheat-treated Eastman membranes. Total run time for this test was 523 1/2 hours.

During the run there were temperature fluctuations as large as 2.5°C between successive data points which resulted in flux variations of as much as 5% from the changes in viscosity. The data shown in Figure 3 are the values actually measured. Flux decline parameters, b, ($b = -\Delta \log \text{flux}/\Delta \log \text{time}$) in this report were determined from plots where the flux data had been normalized to 25°C by assuming that flux is inversely proportional to viscosity. The irregularities seen in the figures, especially between 200 and 350 hours, were almost removed in the normalized plots.

For the heat-treated Eastman membranes operated at 17.6 and 10 ft/sec velocities the flux decline was mild for the first 430 hours (b = 0.030 and 0.017, respectively) with fluxes going from 4.94 to 4.30 gfd and from 3.40 to 3.20 gfd, respectively, between 1.3 and 430 hours of sewage circulation. The rate of decline then increased (b = 0.79 and 0.52) so that the flux was 3.55 and 2.82 gfd after 621 hours of operation. Rejections for these membranes increased with time from 97-94% to greater than 99%. The membrane operated at 3.1 ft/sec axial velocity also showed a slight drop in flux (b = 0.040) from 3.8 to 3.06 gfd during the initial 141 hours of the test. Then a steep decline (b = 1.11) was observed during the remainder of the run with the flux being only 0.733 gfd after 621 hours. The rejection increased during the initial period from 89 to 98% and then decreased slightly to 97% as the fouling layer increased in thickness.

For the Aerojet membranes flux behaviors for membranes at 17.6 and 10 ft/sec were similar to each other during the first 52 hours with flux decreasing from 37 to 32 gfd and from 38 to 33 gfd, respectively, (b = 0.043). The rate of decline then increased slightly (b = 0.17 and 0.33, respectively) until at about 350 hours when severe flux declines started (b = 1.5 and 1.6). After 621 hours fluxes were only 10.1 and 7.6 gfd, respectively. Rejections increased slightly during the test (from 94 to 95-97%). The membrane at 3.1 ft/sec had a severe flux decline dropping from 18.8 to 0.77 gfd during the test; the flux decline parameter, b, was 0.5. Rejection decreased during the test from 88 to 56%.

For the unheat-treated Eastman membrane operated at 17.6 ft/sec a decrease in flux occurred during the initial 285 hours from 115 to 64.6 gfd (b = 0.13). Then a severe decline (b = 4.1) started with the flux dropping to 8.4 gfd after 621 hours of operation. Rejections increased from 36 to 67% during the period of mild flux decline. Then as the flux dropped rapidly the rejection increased faster so that by the end of the test it was 86%. A dynamic membrane layer had apparently formed at this high velocity. For the membrane at 10 ft/sec velocity the flux decline parameter, b, was 0.13 during the first 70 hours. From 70 to 170 hours there was a substantial flux decline, then for the next 100 hours there was a relatively small flux decline and finally for the last 300 hours another substantial flux decline with b = 2.8 during this final period. Rejection increased slowly during the entire test going from 42 to 55%. A severe flux decline occurred from the start of the run for the membrane operated at 3.1 ft/sec. The flux dropped from 114 gfd at 30 minutes after the start of water circulation to 19.4 gfd after 1.3 hours of circulating sewage (total circulating time of 2.8 hours). After 621 hours of sewage operation the flux was only 0.85 gfd; the flux decline parameter during sewage circulation was 0.5. Rejection decreased slightly during the test, going from 43 to 39%.

After 620 hours of operation ~100 mg/ℓ calcium hypochlorite was added to the primary sewage feed and circulated for a total of two hours (with no depressurization). Almost immediately fluxes from the high flux membranes operating at 10 and 17.6 ft/sec were restored to values corresponding to an extrapolation (on a log flux-linear time plot) of results for the first 300 hours of the run. The moderate rate of flux decline observed during the first 300 hours of the test continued for the next 140 hours after the calcium hypochlorite wash. Additional details about this treatment will be given in a subsequent section titled "Intermittent Application of Chlorine."

Two other tests were run with membranes and axial velocities the same as the previous test (Run 26). These runs are listed in Table 5 as 25 and 28. A plot of log flux vs time for the first ~300 hours of these three runs is shown in Figure 4; results from the previous figure (Run 26) are shown as solid points, results from Run 25 are shown as open points and results from Run 28 are shown as half-filled points. Initial fluxes and rejections 1/2 hour after startup with city water feed are given in Table 5.

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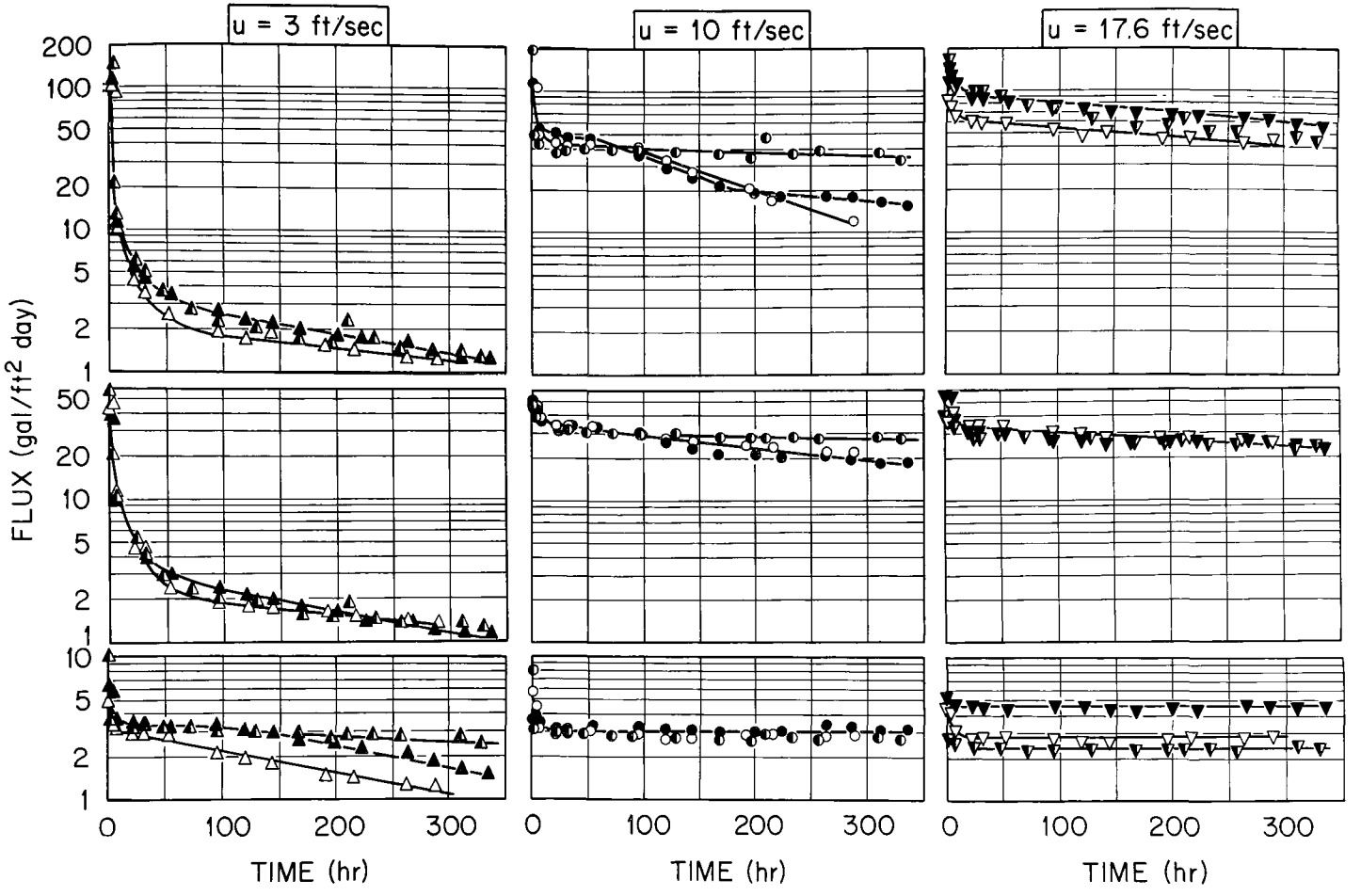

Figure 4. Effect of Flux and Axial Velocity on Flux Decline Using Primary Sewage as Feed. (Cellulose Acetate Membrane, 600 psig, △,○,▽, Run 25; ▲,●,▼, Run 26; ◭,◐,▼, Run 28)

For clarity, the 27 different sets of results from Runs 25, 26, and 28 are separated into nine individual graphs in Figure 4. Graphs in each column are for the same velocity but different membranes (easily identifiable by the megnitude of the flux), while graphs in each row are for the same membrane but different velocities, i.e., the top row in the figure is for the high flux unheat-treated membrane at velocities of 3, 10, and 16 ft/sec.

The results shown in Figure 4 are notable for several reasons. First the rather remarkable agreement of the three different tests. If the fluxes had been normalized to the same initial flux the agreement would have been even better in several cases. The greatest divergence among individual tests in the three runs occurred with the lowest initial flux membranes exposed to a velocity of 3 ft/sec and the highest initial flux membrane exposed to a velocity of 10 ft/sec. As will be shown later the conditions for these two tests place them very close to the "threshold velocity" for prevention of particulate fouling. In other words the deviations within these two particular combinations of initial flux and axial velocity are probably not due to variations in the primary effluent but to the proximity to the threshold velocity.

Second it is clear that the rate of flux decline is a function of both initial flux and the axial velocity. For the high flux membrane, within 50 hours of operation at 3 ft/sec the flux had decreased from over 150 gal./ft^2•day. However flux from the same type membrane exposed to an axial velocity of 17.6 ft/sec was still over 90 gal./ft^2•day after 50 hours and at the end of 300 hours the flux was ~60 gal./ft^2•day. This will be discussed further in a subsequent section of the report.

Effect of Loop Depressurization on Flux Recovery

Run 33. The primary objective of this test was to obtain additional information on the effect of axial velocity and initial flux on the rate of flux decline. However there were three unscheduled loop shutdowns during the test and as a result the most informative aspect of these tests was the flux recovery after the shutdown for the different operating conditions.

Membranes used were Eastman HT-00 unheat-treated, similar material heat-treated at 90°C for 5 minutes, and Aerojet-General 6% membrane. Axial velocities were 3.3, 8, and 21.8 ft/sec. System pressure was 600 psig. Temperature of the circulating fluid ranged from 31.3 to 33.3°C.

City water spiked with NaCl to ~0.01 M was circulated for the first 2 hours to determine the initial flux and rejection of each membrane.

For the heat-treated Eastman membranes initial fluxes were 2.0 to 2.5 gfd with rejections of 92-99% with the highest rejection associated with the highest velocity. For the Aerojet membranes fluxes were 48 to 51 gfd with rejections of 92 to 94%. The unheat-treated Eastman membranes had fluxes of 61.4 to 65.5 gfd with rejections of 46 to 51%.

Flux and rejection as a function of time are shown in Figure 5; separate plots are given for each membrane at each different velocity. As in previous figures the upper row of plots are for the Eastman unheat-treated membranes; the middle row is for Aerojet 6% membranes and the bottom row of plots is for the Eastman membrane heat-treated to 90°C. Fluxes before and after shutdown are given in Table 8.

Table 8. Effect of Loop Shutdown on Flux (Run 33, 600 psig)

Shutdown No.		1		2		3	
Run Time, hr		84		158		286	
Duration		9 1/2 hr		1 min		14 hr	
	Axial	Flux, gal./ft^2•day					
Membrane	Velocity ft/sec	32 hr before	0.8 hr after	1 hr before	6 hr after	9.5 hr before	1 hr after
Eastman Heat-treated	21.8	1.7	1.6	1.6	1.6	1.6	1.6
	8.0	2.2	2.2	2.1	2.2	2.2	2.3
	3.3	1.7	1.7	1.4	1.5	1.5	1.4
Aerojet 6%	21.8	32	31	29	30	26	26
	8.0	26	23	14	14	7	12
	3.3	4.6	11	3.3	3.3	1.8	2.0
Eastman Unheat-treated	21.8	42	42	38	39	39	36
	8.0	25	27	14	13	7.1	11
	3.3	5.1	11	3.5	3.5	1.9	2.1

The first shutdown (of 9.5 hours duration) occurred after 84 hours of sewage circulation when the city power failed during a thunderstorm. When the loop was restarted, there was no significant change in flux for six of the membranes, a small increase for one membrane and appreciable increases in flux for two membranes. The greatest increase was observed with the two higher flux membranes exposed to the lowest velocity, well below what we now know is the threshold velocity for these membranes. However, at the time of the shutdown, flux from both membranes had declined from an initial value of ~50 gal./ft^2•day to ~4 gal./ft^2•day and since the flux after startup was only ~10 gal./ft^2•day the recovery is largely of academic interest.

The second outage of the test, caused by a momentary power failure during an electrical storm, occurred after 158 hours of sewage circulation and lasted only about 1 minute. This shutdown had little or no effect on the flux.

The third shutdown, also caused by a power failure during a storm, occurred after 286 hours of sewage circulation and lasted 14 hours. This shutdown had no effect on the lowest flux membranes at all three velocities or on the two higher flux membranes at the highest velocity, 21.8 ft/sec.

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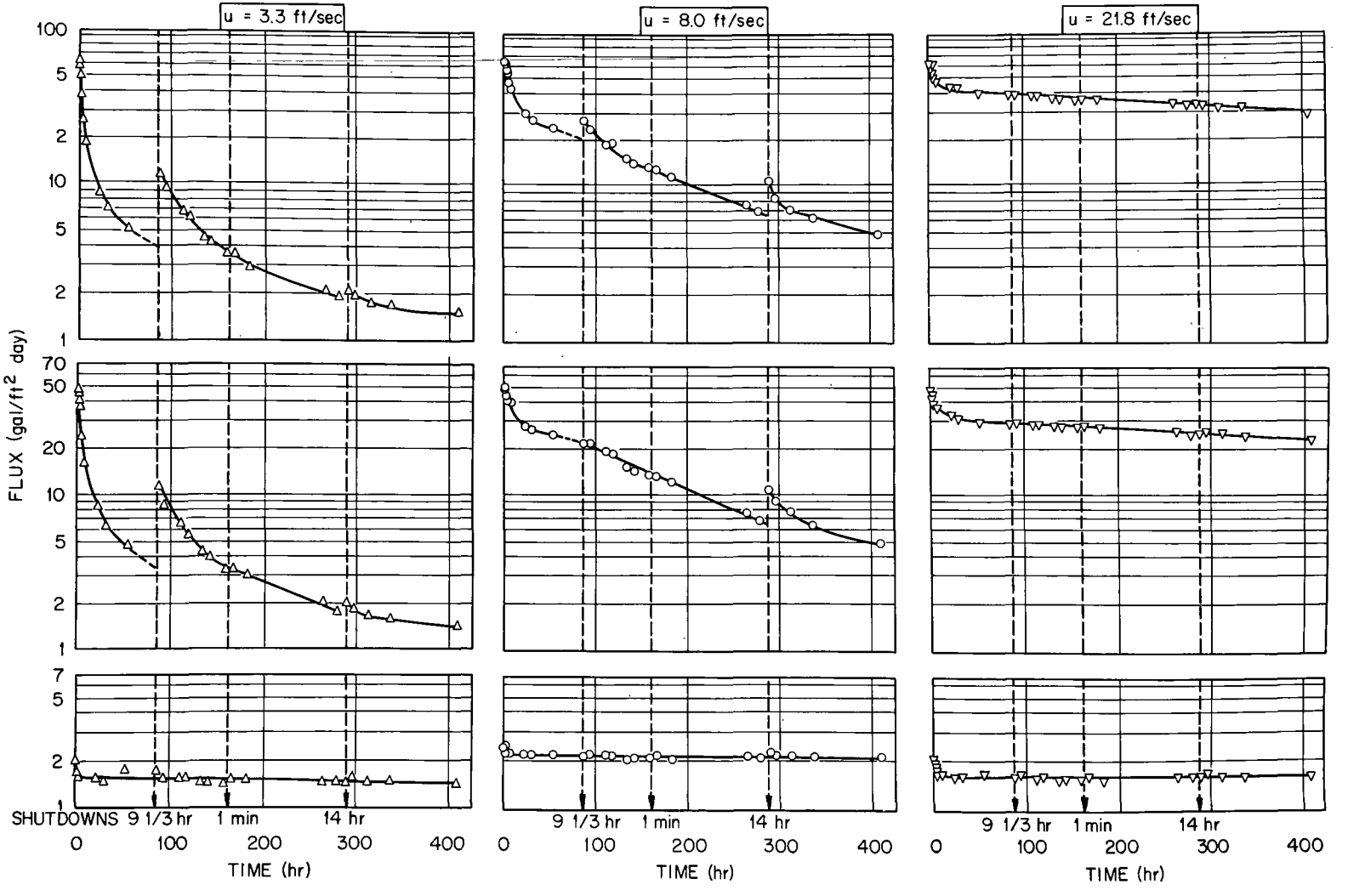

Figure 5. Flux Recovery After Shutdowns as a Function of Axial Velocity and Initial Membrane Flux. (Primary Sewage Feed, 600 psig, Cellulose Acetate Membrane, Run 33)

The greatest effect was observed with the two higher flux membranes at the intermediate velocity, 8.0 ft/sec. With both membranes the flux had declined from ~60 gal./ft^2•day to ~7 gal./ft^2•day before the shutdown. After the shutdown the flux increased to ~11 gal./ft^2•day, again an amount which is largely of academic interest.

From the results of this and other tests we believe that the principal effect of a depressurization is to loosen heavy layers of particulate fouling sufficiently that at least a portion of them can be flushed from the system. Because the Aerojet and Eastman unheat-treated membranes at 3.3 ft/sec had such a low flux after the first shutdown, the axial velocity was sufficiently near "threshold" that little additional particulate fouling occurred. Hence subsequent shutdowns had little effect on their flux.

Intermittent Application of Chlorine

Run 26 (Continued). In Run 26 membranes with initial fluxes of ~120, ~50, and ~5 gal./ft^2•day were exposed to primary sewage feed at axial velocities of 3.1, 10, and 17.6 ft/sec. During the first 300 hours the flux decline of the two higher initial flux membranes at the higher axial velocities appeared to stabilize at a rather modest value. However, from ~300 hours to 600 hours a severe flux decline occurred with these membranes (see Figure 3).

After 622 hours of sewage circulation, HTH, a commercial chemical compound containing 70% calcium hypochlorite, was mixed with the primary sewage feed stream to give a concentration of about 100 mg/ℓ calcium hypochlorite and was used as the feed for about one hour. This was done to determine whether part of the deposit that had formed on the loop surfaces could be removed. The treatment was repeated four hours later. The immediate effects on the flux are summarized in Table 9 which shows flux data taken 45 minutes before the start of the treatment and those taken one hour after it was completed. Longer-term results are shown in Figure 6 which is a plot of flux as a function of total run time with the time of the calcium hypochlorite addition indicated by a dashed line and the notation "add HTH."

The effectiveness of the treatment of the flux appeared to be velocity dependent. Little or no change occurred for membranes operated at 3 ft/sec velocity. For membranes at 10 ft/sec velocity the flux increased by 28 to 75%. For membranes at 17.6 ft/sec the flux increased 36 to 317%. Rejections decreased only a few per cent except for the unheat-treated Eastman membrane operated at 17.6 ft/sec which dropped from 86.4 to 65.7%. This membrane had the greatest flux increase, from 8.39 to 35.0 gfd.

The run was continued for about 140 hours to observe the effect the calcium hypochlorite treatment had on the flux decline. During this period the flux decline was mild for all nine membranes as can be seen in Figure 6. For the Aerojet and the unheat-treated Eastman membranes at the two higher velocities the severe flux declines observed during

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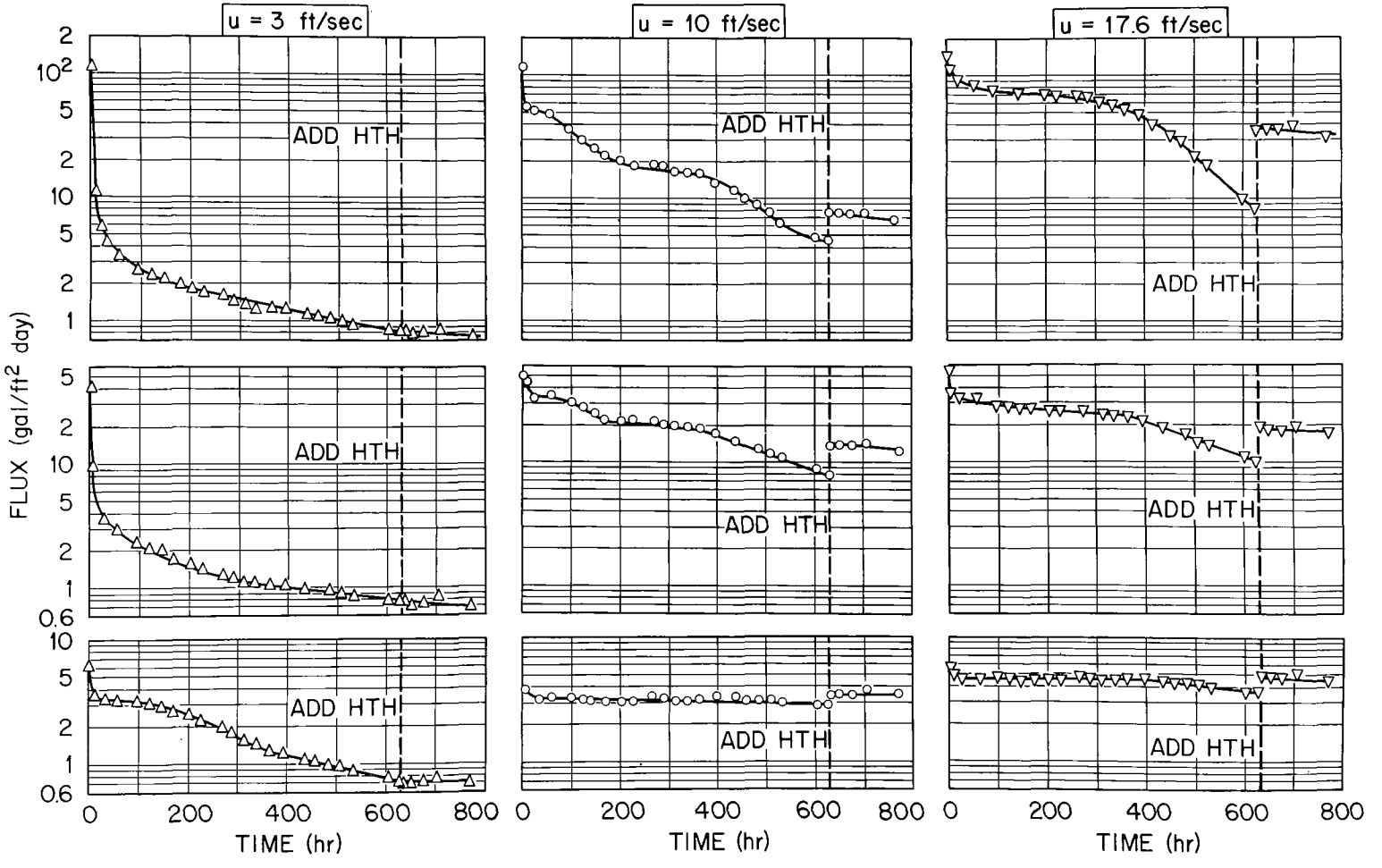

Figure 6. Effect of Flux and Axial Velocity on Flux Decline Using Primary Sewage as Feed. (Cellulose Acetate Membrane, 600 psig, Run 26)

Table 9. Effect of Calcium Hypochlorite Wash on Flux (Run 26, 600 psig, CaCl(OCl) Added After 624 Hours Total Run Time)

	Flux, gfd			
Velocity ft/sec	Initial	45 Minutes Before Wash	One Hour After Wash	140 Hours After Wash
3.1	114	0.85	0.85	0.76
	39	0.77	0.76	0.71
	6	0.73	0.73	0.72
10	116	4.6	7.0	6.5
	49	7.6	13.3	12.1
	4	2.8	3.3	3.3
17.6	144	8.4	35.0	29.9
	56	10.1	18.3	16.6

the 200 hours preceding the treatment did not occur but rather the declines were similar to those which occurred between 50 and 400 hours of circulation. The fluxes after the calcium hypochlorite wash had recovered back to the extrapolated flux from the 50 to 400 hour portion of the run.

Figure 7 shows the membranes after they were removed from the test sections at the conclusion of the run. Membranes from the low velocity test section, shown on the left side of the figure, were coated with a heavy, soft sludge. The quantity of deposited material decreased as the axial velocity increased. The unheat-treated Eastman membrane at 17.6 ft/sec velocity had essentially no solids on the surface. Note that the membrane with an initial flux of ~50 gal./ft^2•day exposed at an axial velocity of 10 ft/sec was only partially covered with a continuous deposit; the uncoated part of this membrane, however, was speckled with small deposits of sewage particulates elongated in the direction of flow. An enlarged photograph of this membrane is shown in Figure 8. The appearance of this membrane suggests that some sort of nucleation phenomena may play a role in the adherence of particulate solids to cellulose acetate membranes.

Run 28. The objective of this test was to determine whether addition of calcium hypochlorite to the primary feed at approximately 250-hour intervals would prevent the severe flux decline which was observed after ~400 hours of operation in Run 26.

Membranes used in this test were Eastman HT-00 unheat-treated (J_I = initial flux ~150 gfd), the same type membrane heat-treated at 90°C ($J_I \sim 5$ gfd), and Aerojet-General 6% ($J_I \sim 50$ gfd). System pressure was maintained at 600 psig. Temperature of the circulating fluid ranged between 25.5 and 31.8°C. Axial velocities in the test sections were 3.1, 10, and 17.6 ft/sec. Total run time was 983 hours; two shutdowns occurred one after 209 hours with a duration of 7 1/2 hours, the other after 890 hours with a duration of 14 2/3 hours.

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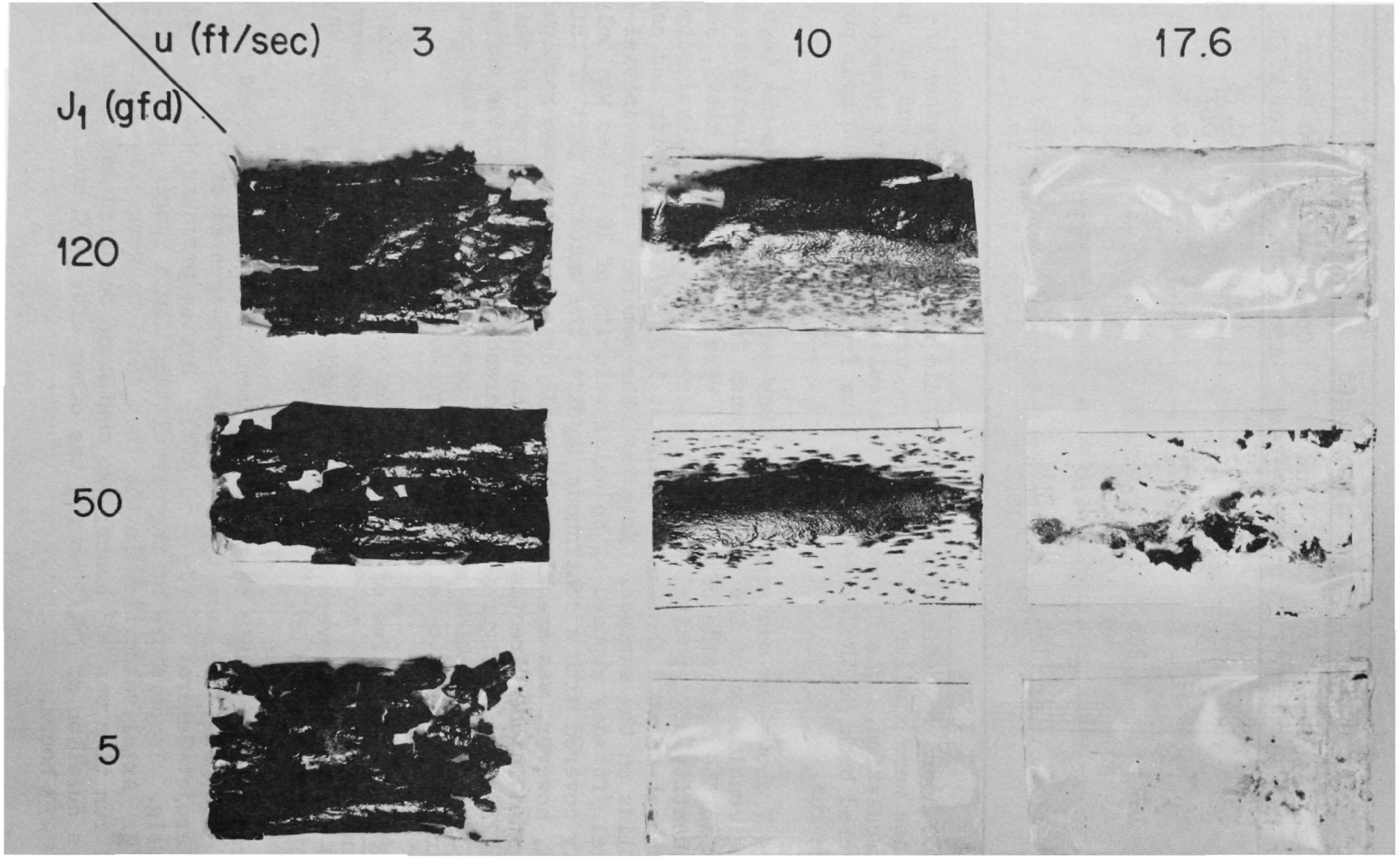

Figure 7. Membranes from Run 26 After 768 Hours Exposure to Primary Sewage Feed.

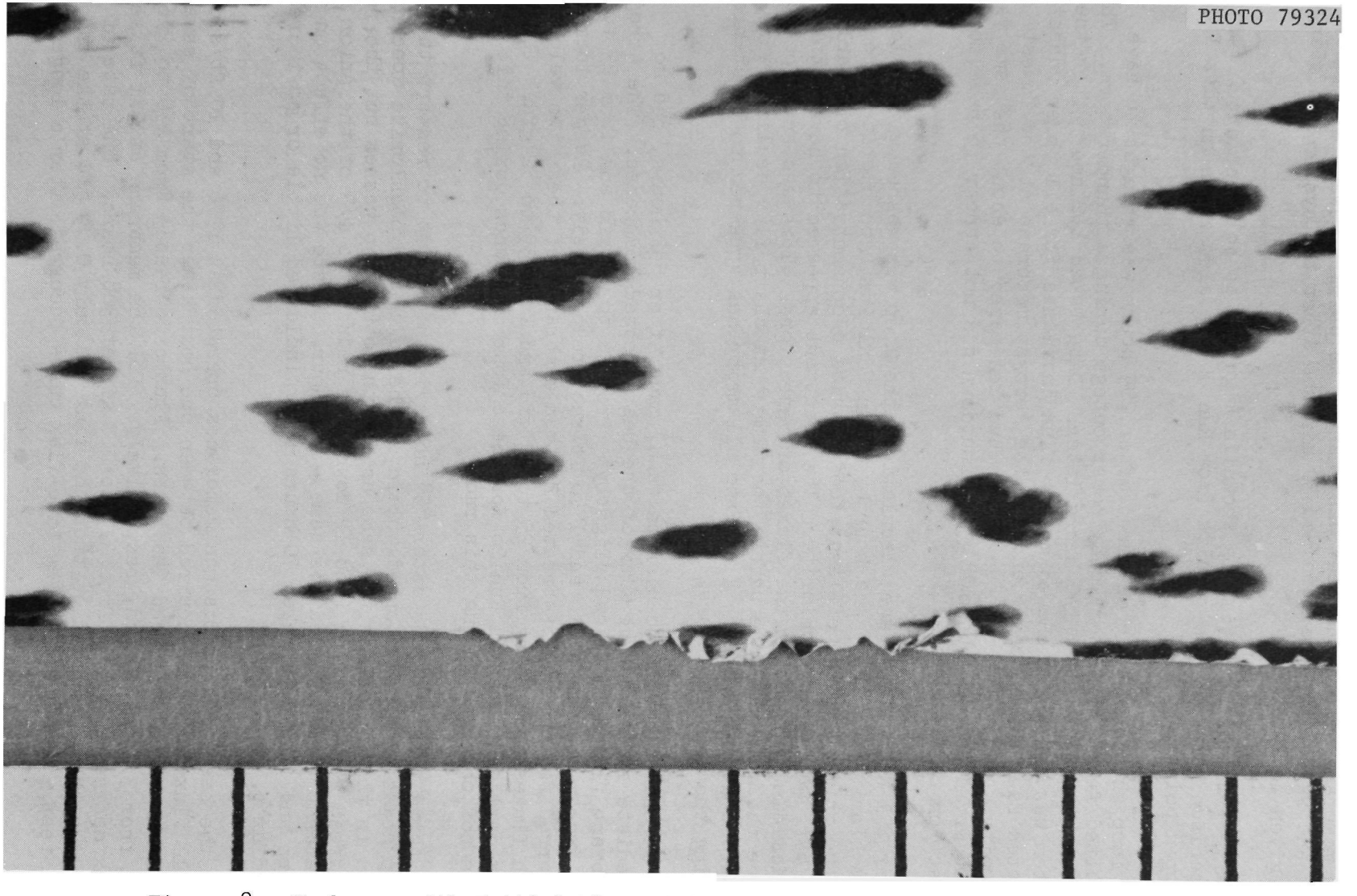

Figure 8. Membrane with Initial Flux of 50 gfd Exposed to Primary Sewage Effluent Feed for 768 Hours at an Axial Velocity of 10 ft/sec. (Run 26, 600 psig, smallest scale division = 1/16-in.)

City water spiked with NaCl to ~0.01 M was circulated for the first 2 1/4 hours of the run to determine the initial flux and rejection for each membrane before the feed was changed to primary sewage effluent. Flux and rejection data obtained 30 minutes after the system was started are given in Table 5.

Flux as a function of time is shown in Figure 9 as solid points. For comparison purposes the results from Run 26 are also shown on this figure as open points.

The loop shutdown which occurred after 209 hours was thought to have resulted from a momentary city power outage during a thunderstorm. The system was restarted 7 1/2 hours later. Data taken 1/2 hour after the system was restarted showed modest flux increases for all three unheat-treated Eastman membranes and for the Aerojet membrane at the lowest velocity. A day later these fluxes had decreased to about the same values as observed before shutdown. The shutdown did not appear to have much effect on membrane rejection.

After 231 hours of sewage circulation, the loop was washed with calcium hypochlorite for about one hour by adding calcium hypochlorite directly to the primary sewage effluent feed to give a concentration of nearly 500 mg/ℓ calcium hypochlorite. The treatment appeared to have little or no immediate effect on the fluxes or rejections. This was anticipated since the wash was made before the time that sharp flux declines were observed during Run 26. Flux from all membranes appeared to be stable during the next 200 hours.

The second calcium hypochlorite wash, made after 542 hours, had no immediate effect on the flux of the heat-treated membranes. The Aerojet membrane fluxes increased slightly – from 26 to 28 gfd and from 23 to 25 gfd from membranes at 10 and 17.6 ft/sec, respectively. Larger increases occurred for the unheat-treated Eastman membranes at these two velocities with fluxes increasing from 25 to 32 gfd and from 39 to 52 gfd. The sharp flux declines noted in Run 26 for these four membranes during this period were not observed during this run.

The third wash was made after 758 hours in an attempt to restart two feed pump heads that had stopped working. The calcium hypochlorite concentration in the system was the same as for the other two washes but the rate of fresh feed addition (and letdown) was reduced to 1/3 of the prior rate. Treatment time remained the same – one hour. There was no effect on the pumps and flux data taken 70 hours later indicated little or no increase in fluxes.

For the heat-treated Eastman membranes operated at 17.6 and 10 ft/sec very small flux declines occurred between one hour after the start of sewage effluent circulation and 945 hours. Fluxes decreased from 2.8 to 2.6 gfd and from 3.1 to 3.0 gfd, respectively. For the membrane at 3.1 ft/sec the flux decreased slowly from 3.7 to 2.7 gfd at 308 hours. The rate of decline then increased so that the flux was only 0.89 gfd at the end of the test. The break in the time-flux curve occurred after a longer time

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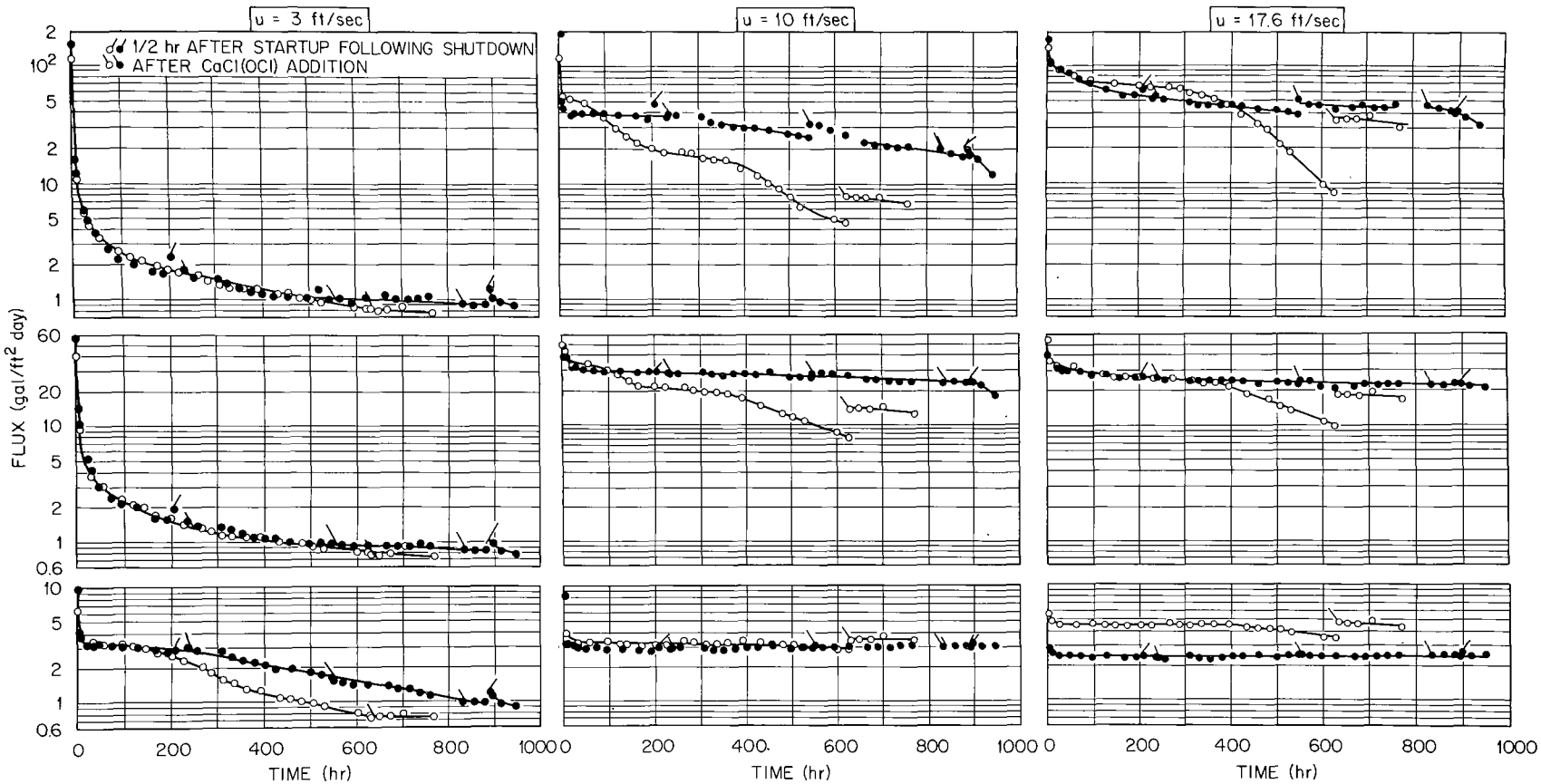

Figure 9. Comparison of Effect of Adding CaCl(OCl) Before Severe Fouling Occurs on all Membranes (Solid Points) with Addition After Severe Fouling Occurs (Open Points). (Primary Sewage Feed, 600 psig, Cellulose Acetate Membranes, Solid points - Run 28, Open points - Run 26)

period than in Run 26. Rejections increased slightly with time for all of the membranes going from 93-96% to >98%.

Calcium hypochlorite treatment had no effect on the intermediate and high flux membrane exposed at an axial velocity of 3 ft/sec. However, at axial velocities of 10 and 17.6 ft/sec there was a dramatic effect of the intermittent chlorine addition on the flux from both the intermittent and high initial flux membranes. After the initial flux decline in the first 30-40 hours flux decline was stabilized at a very small value for the remainder of the run by the chlorine additions at 231, 542, and 758 hours. With the Aerojet membranes, chloride rejections were better than 95% for the entire 950 hours indicating that, at least for this period of time, the brief exposures to chlorine did not damage the rejection capability of the membranes.

Although the amount of chlorine, the length of time between flushes and the duration of flushes was not optimized in these studies, it is clear that unless the axial velocity is above the threshold velocity for particulate fouling the use of an intermittent chlorine flush is totally ineffective in restoring flux. With proper hydrodynamics, intermittent chlorine flushes, at least in this one test, prevented an expected catastrophic flux decline in the period from 400 to 950 hours run time.

Modification of Membrane Characteristics by Additives

Several exploratory studies have indicated that modification of membrane characteristics (i.e., charge on the membrane) may offer protection against fouling since most polymers in sewage are probably anionic in nature.

Kraus (5) reported qualitative observations supporting this hypothesis in studies of the application of dynamic membranes to treatment of sewage. Specifically it was noted in those studies that although rejections of cation-exchange membranes were decreased on exposure to sewage, the decrease was not as serious as that observed on exposure of anion-exchange membranes to sewage.

The Environgenics Company (10) developed procedures for attaching enzymes to cellulose acetate membranes. Although enzyme activity was markedly reduced in the process, the activity remaining was significantly stabilized by attachment of the enzyme to the membrane. No tests were reported of the characteristics of this membrane in the presence of sewage. However, the cellulose acetate hydrogen succinate (CAHS) used in preparing the enzyme membrane was tested in contact with secondary effluent. The first six days of the test indicated no change in flux while rejection increased from 80 to 87%. From 6 to 8 days the flux decreased from 15.5 to 13.5 gfd and then to 6 gfd after 16 days. The complete absence of flux decline in the first 6 days was taken to be an indication that the negatively charged CAHS may have had an inherent fouling resistance.

Keilin (14) reported promising results in improving salt rejection with polyvinylmethyl ether (PVME). Interestingly, the additive was reported

to increase rejection with only a slight reduction of the flux. However, the effects of the PVME decayed over a period of about one day after the ether was flushed from the system. Also, it was reported (15) that PVME will precipitate from dilute solutions at a temperature above about 33°C.

A secondary objective of the present program was to determine the effect on flux and rejection, if any, of a pretreatment of the membrane with additives. Two additives were tested: polyvinylmethylether and polyacrylicacid; the first with the objective of increasing the rejection of the unheat-treated high flux membrane, the second with the objective of developing a charge on the membrane to minimize adherence of foulants. The results of each test will be described individually.

Run 19. Axial velocities for this test were 5.2, 9.9, and 15.3 ft/sec, feed was primary sewage effluent. Temperature of the circulating fluid varied between 30 and 32.5°C and the system pressure was maintained at 600 psig.

During the first 19 hours a 0.01 M solution of NaCl containing 100 mg/ℓ. PVME was circulated for pretreatment with the letdown and product streams being returned to the feed. During the pretreatment the flux from the Aerojet membranes decreased (at all velocities) from 37-49 to 26-28 gfd and rejection increased from 77-79 to 95-97%. The flux and rejection for the Eastman membrane, operated at an axial velocity of 15.3 ft/sec, remained constant at about 120 gfd and 1%, respectively. At 9.9 and 5.2 ft/sec velocities, the rejection increased from a 31-45% range to 78-81% range while the fluxes decreased from 80-87 to 23-35 gfd. Most of the change in the rejections occurred during the first two hours of circulation. Although the increased rejection is of interest, the fact that the flux declined markedly at the same time means that the result is of little practical interest.

Because flux through cellulose acetate membranes varies with applied pressure and viscosity, measured fluxes for this test were corrected for small variations in temperature and pressure by the relation

$$J_s = J\left(\frac{600}{P}\right)\left(\frac{\mu_T}{\mu_{30°}}\right) ,$$

where J_s = normalized flux,

J = measured flux,

P = system pressure,

μ_T = viscosity of water at system temperature, and

$\mu_{30°}$ = viscosity of water at 30°C.

For the Aerojet membranes only modest flux declines were observed during the 215-hour run at velocities of 9.9 and 15.3 ft/sec (from 26 to 22 gfd and from 26 to 21 gfd, respectively). Rejections were greater than 87%. There appears to be a slight offset in the flux decline curve at ~35 hours

particularly for the results from the membrane operating at 9.9 ft/sec. At a velocity of 5.2 ft/sec a severe flux decline occurred with this membrane, from 27 to 4.5 gfd, while the rejection remained fairly constant at 97%.

For the Eastman membrane at a velocity of 9.9 ft/sec, flux increased from 36 to 43 gfd during the first 30 hours on sewage and then decreased to 24 gfd by the end of the run. Rejection decreased from 83 to 41% during the first 30 hours and to 34% by the end of the test. At a velocity of 15.3 ft/sec, flux decreased from 128 to 80 gfd during the 215-hour run with little or no discontinuity at 30 hours after addition of primary sewage (in contrast with the bahavior at 9.9 ft/sec). Rejections were less than 20% for the entire run. At an axial velocity of 5.2 ft/sec there was a severe flux decline during the run, from 24 to 2.6 gfd and rejection decreased from 90 to 54%.

It is interesting to note that of the six different combinations of initial flux and axial velocity, only the results from the membrane with an initial flux of ~80 gfd and axial velocity of 9.9 ft/sec resembles the brackish water results of Keilin (14). That is, the flux increase from 31 to 45 gfd and rejection decrease from 83 to 41% in the first 30 hours apparently corresponds to Keilin's observation that the effect of the PVME disappeared ~1 day after the PVME was flushed from the system. Apparently with primary sewage as feed there is an optimum velocity (not necessarily 9.9 ft/sec) required to obtain the benefit of the PVME membrane treatment. In none of the tests was there a negligible effect of PVME on flux.

Run 20. All membranes used in this test were soaked for 65 hours in water containing 100 mg/ℓ PVME prior to installation on the test unit. The temperature during pretreatment was maintained below 30°C to prevent precipitation of the PVME. This pretreatment procedure was used to permit the ether to diffuse into the membrane and perhaps prolong the effect of the PVME.

Axial velocities were 4.75, 9, and 12 ft/sec; the temperature of the circulating fluid varied between 28.5 and 31.5°C, and system pressure was 600 psig. City water spiked with NaCl (~0.01 M) was circulated for the first 18 hours. During this time fluxes for the Aerojet membranes remained essentially constant at 40 to 60 gfd except for the last measurement at the 9 ft/sec velocity which showed an increase to 182 gfd. The rejection for this membrane dropped from 77 to 16% indicating that a defect had developed in the membrane. Rejection for the membrane at 12 ft/sec was constant at 92% while at a velocity of 4.75 ft/sec it decreased from 89 to 58%. For the Eastman membranes flux was about 180 gfd at velocities of 9 and 12 ft/sec while it decreased from 184 to 86 gfd at 4.75 ft/sec. Rejections of the Eastman membrane was less than 15% at all axial velocities.

After changing to primary sewage mild flux declines were observed for the Aerojet membranes at 9 and 12 ft/sec during the next 215 hours (37 to 27

gfd at 9 ft/sec and 39 to 33 gfd at 12 ft/sec). At 4.75 ft/sec, flux dropped from 36 to 2.6 gfd during the 215-hour test. At the two higher velocities rejections ranged from 92 to 97%. At the low velocity it decreased from 92 to 66%.

For the Eastman membrane a severe flux decline occurred at 4.75 ft/sec (from 36 to 2 gfd in 215 hours). At an axial velocity of 9.9 ft/sec there was a mild decline during the first 79 hours on sewage followed by a sharper decline to the 215-hour point (from 81 to 44 gfd and finally to 20 gfd). The flux data from the membrane at 12 ft/sec velocity were scattered with higher fluxes measured in the mornings than in the afternoons. Turbidity of the feed did not show this type of variation. During the 215-hour period the flux varied between 29 and 48 gfd with an increasing trend. Rejections remained less than 30% during this period for all Eastman membranes.

There were three flow interruptions after 215 hours of sewage circulation and before the conclusion of the test at 322 hours. In general fluxes were higher after a startup than prior to the stoppage.

Comparison of the results from the run in which the PVME was circulated past the membrane for 18 hours with results from the run in which the membrane was soaked for 65 hours in a PVME solution suggests that circulation of the pretreating solution is necessary for the development of appreciable rejection by the unheat-treated membrane. At 10 ft/sec circulation velocity, pretreatment of the unheat-treated membrane with the circulating PVME solution for 18 hours developed a rejection of ~80% for the nominally zero rejection membrane. (However the flux decreased from 80 to 23 gfd in contrast to Keilin's observation of negligible effect of PVME on flux.) This rejection decayed to ~41% in the first 30 hours after PVME treatment and the flux increased slightly from 36 to 43 gfd during this same period. Soaking the unheat-treated Eastman membrane in PVME had little effect on the rejection for any of the velocities tested.

Run 24. The object of this run was to investigate the effect on flux and rejection of long-term soaking of membranes in polyacrylicacid (PAA) a negatively charged polymer and to determine the change of flux with time for circulation periods in excess of the normal 200-hour test. Membranes used for this test were Aerojet-General 6% and Eastman HT-00. Membranes were secured to the test fingers and placed in a solution containing 100 ppm PAA for 31 days.

Axial velocities for this test were 5.2, 9.9, and 15 ft/sec. Temperature of the circulating fluid ranged from 22.5 to 27°C. System pressure was maintained at 600 psig. However, some pressure transients, < 5 seconds duration, occurred during the run. System pressure would drop 200 psi, then rise 250 psi, and finally return to normal. The transients were caused by erratic operation of check valves in the feed pump.

City water spiked with NaCl (~0.01 M) was circulated for the first 2 1/2 hours. Reference fluxes for the Aerojet membranes ranged from 47 to 40 gfd with rejections of 79 to 93%. The membrane having the highest flux

had the lowest rejection. For the Eastman membranes initial fluxes varied from 122 to 147 gfd with rejections of 10 to 19%. Flux and rejection increased with increasing axial velocity. The PAA appeared to have little or no effect on initial flux or rejection for either membrane.

Primary sewage effluent was circulated for a total of 752 hours. Two flow interruptions occurred during this period. A 14-hour shutdown occurred after 296.5 hours of circulation when the fuses in the air compressor motor circuit opened. Then a second shutdown (45 minutes duration) was caused by one of the pressure excursions tripping the low pressure cutoff switch.

For the Aerojet membrane at 15 ft/sec velocity the flux decline was small (b = 0.055) for the initial period with the flux decreasing from 40 to 28 gfd between 1 and 283 hours, just prior to the first loop shutdown. The membrane at 9.9 ft/sec velocity also had a mild flux decline (b = 0.083) during the first 187 hours (41.5 to 30 gfd) and then the flux decreased to 25 gfd before the shutdown. Rejection increased slightly with time for both membranes (95 to 96% and 92 to 95%, respectively). For the membrane at 5.2 ft/sec the flux dropped sharply (b = 0.565) from 40 to 1.7 gfd before the shutdown. As the flux decreased the rejection dropped from 94 to 70% presumably due to increased concentration polarization as the thickness of the foulant layer increased with time.

Eastman membranes at 15 and 9.9 ft/sec velocities showed discontinuities in the slope of the flux-time curves at about 187 hours. At 15 ft/sec flux decreased from 88 to 70 gfd during this period (b = 0.060) and then to 57 before shutdown. Rejection remained nearly constant at about 40%. At 9.9 ft/sec the rate was larger (b = 0.13) with flux dropping from 40 to 28 gfd in the first 187 hours and then to 21 gfd before shutdown. Rejection decreased with time from 42 to 34%. A severe flux decline (b = 0.565 occurred for the membrane at 5.2 ft/sec from 31 to 1.7 gfd just before shutdown). Rejection decreased from 50 to 38% during this period.

Based on the combined results of other runs without additives the PAA soak did not appear to have an effect on either the flux or rejection for either membrane during this 287-hour time period. Since the membrane had been soaked in PAA for 30 days prior to the test, any beneficial effect of the PAA was expected to disappear ~30 days after exposure to circulating feed as the PAA diffused out of the membrane.

Additives for Sewage Floc-Size Control

The results of many tests made with primary sewage effluent from the Oak Ridge East Sewage Plant showed that for each type of cellulose acetate membrane tested there was a critical or threshold axial velocity below which severe flux declines occurred (see Correlation Section, this report). The threshold velocity is a function of the initial flux, J_I (J_I = flux 30 minutes after pressurization with city water feed). The threshold velocity may also be a function of the floc size in the circulating fluid.

Therefore, to study the effect of floc size on threshold velocity and on the flux decline parameter, b, flocculating agents must be added to the feed stream.

Jar tests were made using several additives to observe the type of floc formed and to determine the required concentration. Based on the results of the jar tests five runs were attempted with different additives: ferric chloride, Cat Floc (a commercial organic cationic polymer used in sewage treatment), alum, Aqua Nuchar A and Ferri Floc [commercial $Fe_2(SO_4)_3$]. Because of experimental difficulties with the addition system, addition of flocculant was often intermittent and tests were often of short duration. Results of the different tests will be briefly reviewed in the following section.

Run 29. The objective of this run was to determine the effect on flux and rejection of the addition of 70 mg/ℓ Fe as ferric chloride ($FeCl_3$) to a primary effluent feed. Membranes used were Eastman HT-00 unheat-treated, similar material heat-treated at 90°C, and Aerojet 6%. Axial velocities in the test sections were 3.1, 10, and 17.6 ft/sec. System pressure was maintained at 600 psig. Temperature of the circulating fluid varied from 31.5 to 33.5°C.

During the initial part of the test while using city water feed only NaCl was added to the loop. When the feed was switched to primary sewage effluent, NaCl and $FeCl_3$ were added. This was done by pumping a 2 M NaCl and 0.2 M $FeCl_3$ solution into the circulating stream at a rate of about one liter per hour. This concentrated solution (pH 1.2) had a high corrosion rate for the type of stainless steel used in the addition system. A leak developed in the tubing 12 hours after the start of the addition. The addition system was isolated and repaired without depressurizing the loop. After operating for about 35 hours with no additives, $FeCl_3$ solution was again added for about 12 hours before another leak developed. The run was then terminated after a total of 71 2/3 hours of circulation.

Since the flocculant was added for only the first 12 hours and then for another 12 hours later in the run, the flux and rejection data are useful mainly as indicators of the possible effect of floc size during a short period of operation.

Table 11 shows flux and rejection data for each membrane taken after 5 3/4 hours of sewage circulation and data from the previous run (Run 28, which contained no flocculating agent) taken after a similar time period.

The most notable effect occurred with the two higher flux membranes exposed to the lowest axial velocity, where in each case the flux with $FeCl_3$ additive was ~3 times greater after 5 3/4 hours exposure to sewage than in a similar run in which there was no $FeCl_3$ addition.

Table 10. Comparison of Flux and Rejection in Run 29 Using $FeCl_3$ with Flux and Rejection in Run 28 when no Additives Were Used (Primary Effluent Feed, 600 psig Pressure)

Membrane	Axial Velocity ft/sec	Run 29, Flocculant Flux, gfd J_I	Run 29, Flocculant Flux, gfd 5 3/4 Hr on Sewage	Run 29, Flocculant R_{obs}, %	Run 28, No Flocculant Flux, gfd J_I	Run 28, No Flocculant Flux, gfd 5 /34 Hr on Sewage	Run 28, No Flocculant R_{obs}, %
Eastman HT-00 Heat-treated 90°C	3.1	15	3.4	95	10	3.6	96
	10	3.6	3.3	98	8.1	3.1	98
	17.6	4.4	3.2	99	2.8	2.6	99
Aerojet 6%	3.1	77	31	84	59	10	88
	10	57	32	92	49	48	95
	17.6	76	41	95	41	35	95
Eastman HT-00 Unheat-treated	3.1	168	30	28	155	12	48
	10	204	62	20	197	43	33
	17.6	193	87	29	178	108	31

As will be discussed in detail in the "Correlation of Results" section, for primary sewage effluent with no additives the threshold velocity, u_c, (in ft/sec) is related to the initial flux, J_I, (in gal./ft^2•day) by

$$u_c = (1.3 \pm 0.6)(J_I)^{1/2} \quad .$$

The preliminary results given in Table 11 indicate that, by adding a suitable flocculant, it may be possible to reduce the numerical coefficient in the equation above from 1.3 to ~0.5, i.e., for an initial flux of 50 gal./ft^2•day, the threshold velocity may be reduced from ~9.5 ft/sec to ~4 ft/sec.

Run 30. The objective of this test was to determine the effect of Cat Floc (an organic cationic polymer) on the flux decline using primary effluent as feed. Although jar tests indicated that the rate of flocculation with this additive was slower than with $FeCl_3$, the pH of the concentrated additive solution was 7.5 compared with 1.2 for the $FeCl_3$. The higher pH was expected to ease corrosion problems with the addition system. Fluxes at various times during the 192-hour test are given in Table 4. Comparison of fluxes from this test with results from earlier tests with no additive indicated that the Cat Floc had little or no effect on the rate of flux decline. In all probability this is a reflection of the much slower rate of flocculation observed in the jar scale tests.

The test sections were removed at the end of this test before the loop was flushed. The inside of the pressure jacket was coated with thick, dark

deposit, much heavier than was found after 900 hours of operating with no flocculant. The membranes at the lowest velocity were heavily coated with a soft sludge. Membranes at the higher velocities had hard film over parts of the surfaces with nucleation spots for heavier deposits evident. The loop piping also appeared to have a heavier coating of sludge than is normally seen.

Run 31. The objective of this run was to determine the effectiveness of adding 50 mg/ℓ Al in the form of commercial alum [$Al_2(SO_4)_3$ containing 17 wt % Al_2O_3] to the primary effluent feed. Membranes used were Eastman HT-00 unheat-treated (new batch), similar material heat-treated at 90°C for 5 minutes, and Aerojet 6%. Axial velocities in the test sections were 7.4, 12, and 24 ft/sec. System pressure was maintained at 600 psig. Circulating fluid temperature increased from 32.5 at the beginning to 39.5°C at the end of the 58-hour test. This is a much larger increase than normal and indicates solids deposition on the sewage side of the heat exchanger.

Continuous alum additions were started when the feed was switched from city water to primary sewage. Flux and rejections for this test are given in Table 5.

Severe flux declines were observed with both the high flux membranes at all velocities in this test. When the test sections were examined at the end of the run a heavy deposit was observed on all surfaces. The finger that operated at 7.4 ft/sec had the heaviest deposit as is normally observed. The other fingers were also completely coated but the deposit appeared more dense, a probable explanation for the lower fluxes for these membranes. Spectrographic analysis of the deposit indicated that it was primarily aluminum oxide.

Run 32. The object of this run was to test Aqua Nuchar A (100 mg/ℓ) as an additive to the primary effluent feed. Membranes used were Eastman HT-00 unheat-treated, similar material heat-treated at 90°C, and Aerojet 6%. The velocities chosen for this run were 2.9, 6.7, and 19.1 ft/sec. System pressure was maintained at 600 psig. Temperature of the circulating fluid varied from 32.5 to 35°C.

The Aqua Nuchar A addition was started with the start of sewage circulation. It was added to the 2 M NaCl solution that is used for the Cl^- ion addition. The Aqua Nuchar formed heavy flocs in the addition solution which were dense enough to prevent reading the rotameter used to monitor this flow. Flow through the 1/4-in. tubing was not fast enough to prevent the flocs from settling. Within the first hour, flow through the addition system was blocked. The lines were cleaned out without changing the pressure in the circulating system. Flow was restarted but continued for only a short period of time. Further attempts to unplug the lines and maintain flow were unsuccessful. The run was terminated after 47 hours of circulation.

Runs 34 and 35. The objective of these tests was to determine the effect of ~50 mg/ℓ Ferri Floc [commercial $Fe_2(SO_4)_3$] on the rate of flux decline using primary sewage effluent as feed. It was hoped that switching from the chloride to the sulfate would alleviate the severe corrosion problems experienced with the addition system in Run 29. Although there were no piping or tubing failures in these tests, both were terminated by failure of the chemical addition pump diaphragm. Examination of the diaphragm after each test showed that they both failed by a pitting type attack.

Flux and rejections for these two very brief runs are summarized in Table 5. During each test there were equipment failures or addition lines plugged every night so that Ferri Floc addition was not continuous. Despite this, the flux from the membrane with an initial flux of ~50 gfd was at least twice as large as the upper limit defined by the scatter band on Figure 11. This supports our belief that increasing the floc size with a suitable additive has promise as a means for markedly reducing the threshold velocity for particulate fouling. Of course the economic attractiveness of this possibility depends on whether the reduced pumping costs due to lower velocity offset the additional chemical costs. Further studies are required to determine optimum additive and additive concentration for maximum reduction in threshold velocity.

Composite Results

The primary objective of this program was to determine the effect of hydrodynamics on the performance of cellulose acetate membranes. The principal hydrodynamic factors studied were the axial velocity (3 to 30 ft/sec) and the velocity normal to the membrane surface (i.e., the membrane flux) which had a range of from ~0.01 cm/min (~3 gfd) to ~0.4 cm/min (150 gfd). In this section of the report, results from all runs made in the present study are combined into composite figures.

Flux Decline. In prior studies a distinction was often made between an initial rapid flux decline followed by a substantially smaller rate of flux decline. The effect of initial flux and axial velocity on the flux decline in the first 20 hours after exposure of the membrane to primary sewage feed is illustrated in Figure 10. The three parts of the figure show results for membranes with initial fluxes, J_I, (J_I = flux 1/2 hour after startup with city water feed) of ~9, ~46, and ~130 gfd. Because initial fluxes varied somewhat from run to run, the flux after 20 hours exposure to primary sewage feed, J_{20}, was normalized by dividing by the initial flux. The dashed lines on these figures are drawn to include the bulk of the experimental results (Tables 5 and 6) for each set of conditions; as such they define the variability of results due to natural variation of the primary sewage feed.

Above a certain velocity range, which was different for each initial flux, the range of flux declines in the first 20 hours is essentially independent of the axial velocity. For a membrane with an initial flux of ~9 gfd, the threshold velocity is 4 to 5 ft/sec; for an initial flux of ~46 gfd

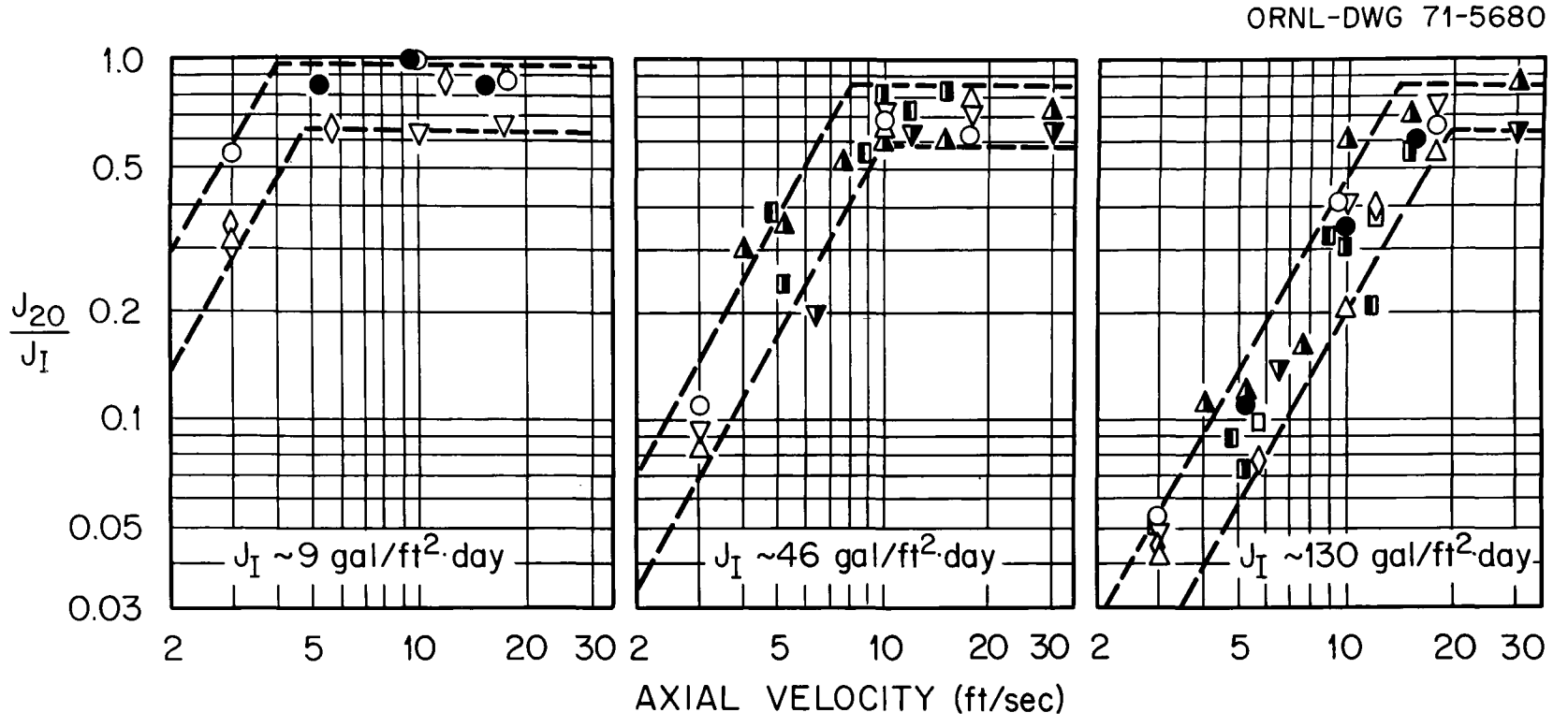

Figure 10. Effect of Axial Velocity and Initial Flux (J_I) on Flux Decline in the First 20 Hours After Exposure to Primary Sewage Feed. (Cellulose Acetate Membrane, 600 psig Pressure)

the threshold velocity is 8 to 10 ft/sec and for an initial flux of ~130 gfd the threshold velocity is 15 to 20 ft/sec.

Below the threshold velocity the range of flux declines in the first 20 hours is strongly dependent on axial velocity. Some initial flux declines in the first 20 hours were so severe that the flux was only 4 to 5% of the initial value.

The extent of the range of flux declines above the threshold velocity was surprisingly similar for the three different initial fluxes; for an initial flux of ~9 gfd, $0.64 < (J_{20}/J_I) < 0.96$; for an initial flux of ~46 gfd, $0.58\ (J_{20}/J_I) < 0.86$ and for an initial flux of ~130 gfd, $0.64 < (J_{20}/J_I) < 0.86$. Additional studies are required to determine factors responsible for the extent of this range and to develop means for insuring that results nearer the upper limit can be achieved routinely.

Flux after 20, 100, and 200 hours exposure to primary sewage feed is shown as a function of axial velocity in Figure 11 for the Aerojet 6% permeation membrane. As in the previous figure there is a threshold velocity above which the axial velocity had little effect on the range of fluxes observed. The threshold velocity for the first 200 hours exposure was substantially independent of exposure time. For this membrane, having an initial flux, J_I, of ~50 gfd, the threshold velocity ranged between 8 and 10 ft/sec. For axial velocities above the threshold there was a definite flux decline during the first 200 hours, from 35 ± 5 gfd at 20 hours to 27 ± 5 gfd at 200 hours. It is notable that this is about the same percentage range of fluxes as in the previous figure.

Membranes exposed at velocities above the threshold showed little or no deposit when removed from the loop. For example, Figure 12 is a photograph of membranes removed from the loop after 750 hours exposure in Run 24. Although the membrane with an initial flux of ~50 gfd exposed to an axial velocity of 9.9 ft/sec was right at the threshold condition, there are only a few speckles on its surface. Nevertheless the flux from this membrane had declined from 47 to 19 gfd during this time. In all probability this flux decline is due to either soluble or colloidal organic material and not to gross particulate fouling.

Figure 13 is similar to Figure 11 except that the results were obtained with a membrane having an initial flux of ~130 gfd. With this membrane the range of threshold velocities was 14 to 20 ft/sec, again substantially independent of exposure time for times up to 200 hours. The flux declined from 90 ± 10 gfd at 20 hours to 60 ± 10 gfd after 200 hours, an average decline of 33% compared to 23% with membranes having an initial flux of ~50 gfd.

Observed Rejections. In previous sections of this report all rejections referred to were chloride rejections. Routinely samples were taken at ~20 and ~200 hours run time with primary sewage feed for more complete analysis. Results for chloride, turbidity, organic carbon, Ca^{++} plus Mg^{++}, inorganic carbon and phosphate are shown in Figures 14 and 15 for the nominal 94% rejecting membrane and in Figures 16 and 17 for the nominal

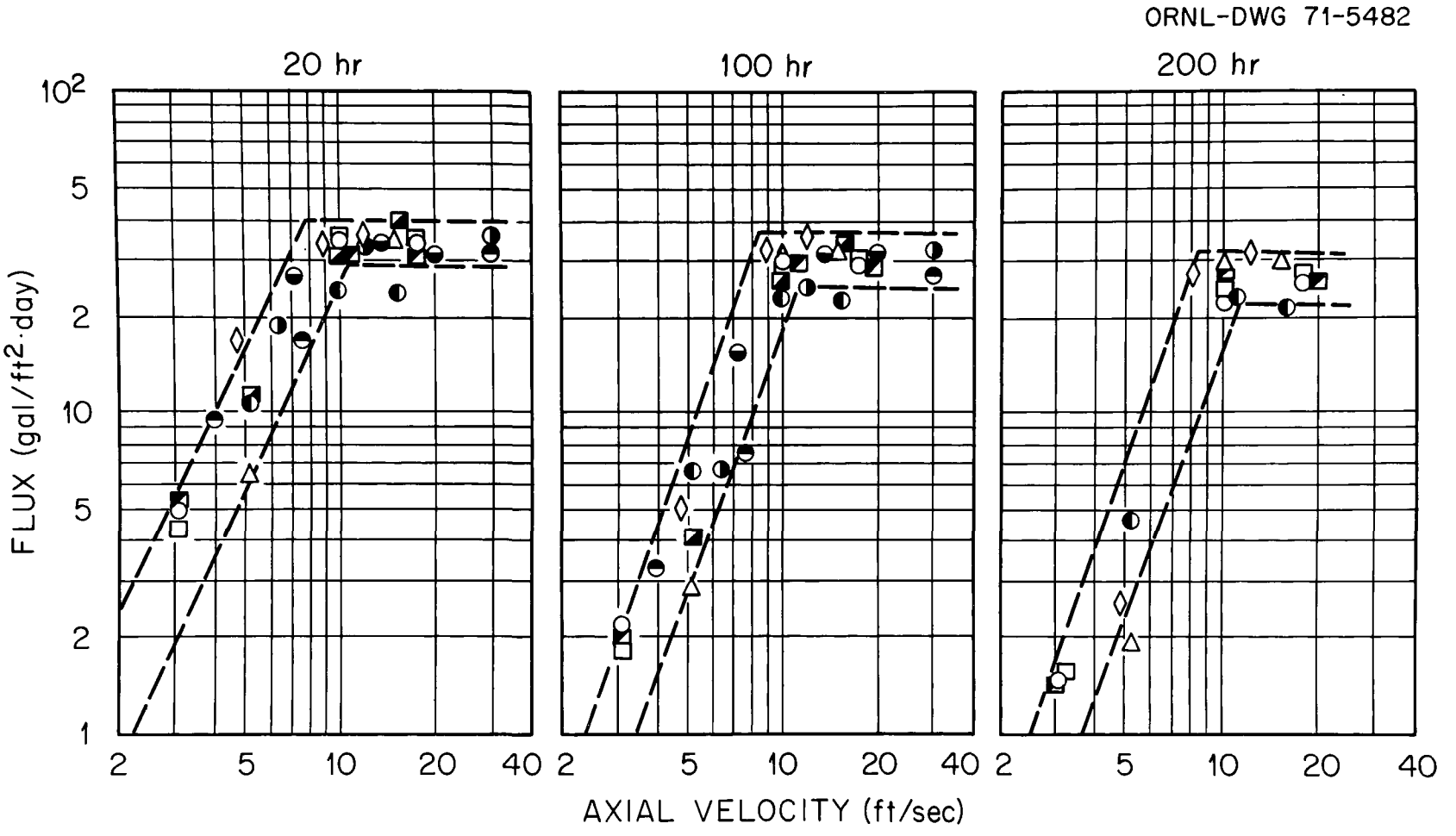

Figure 11. Effect of Axial Velocity and Time on Flux. (Cellulose Acetate, Aerojet 6%, 600 psig, Primary Sewage Feed)

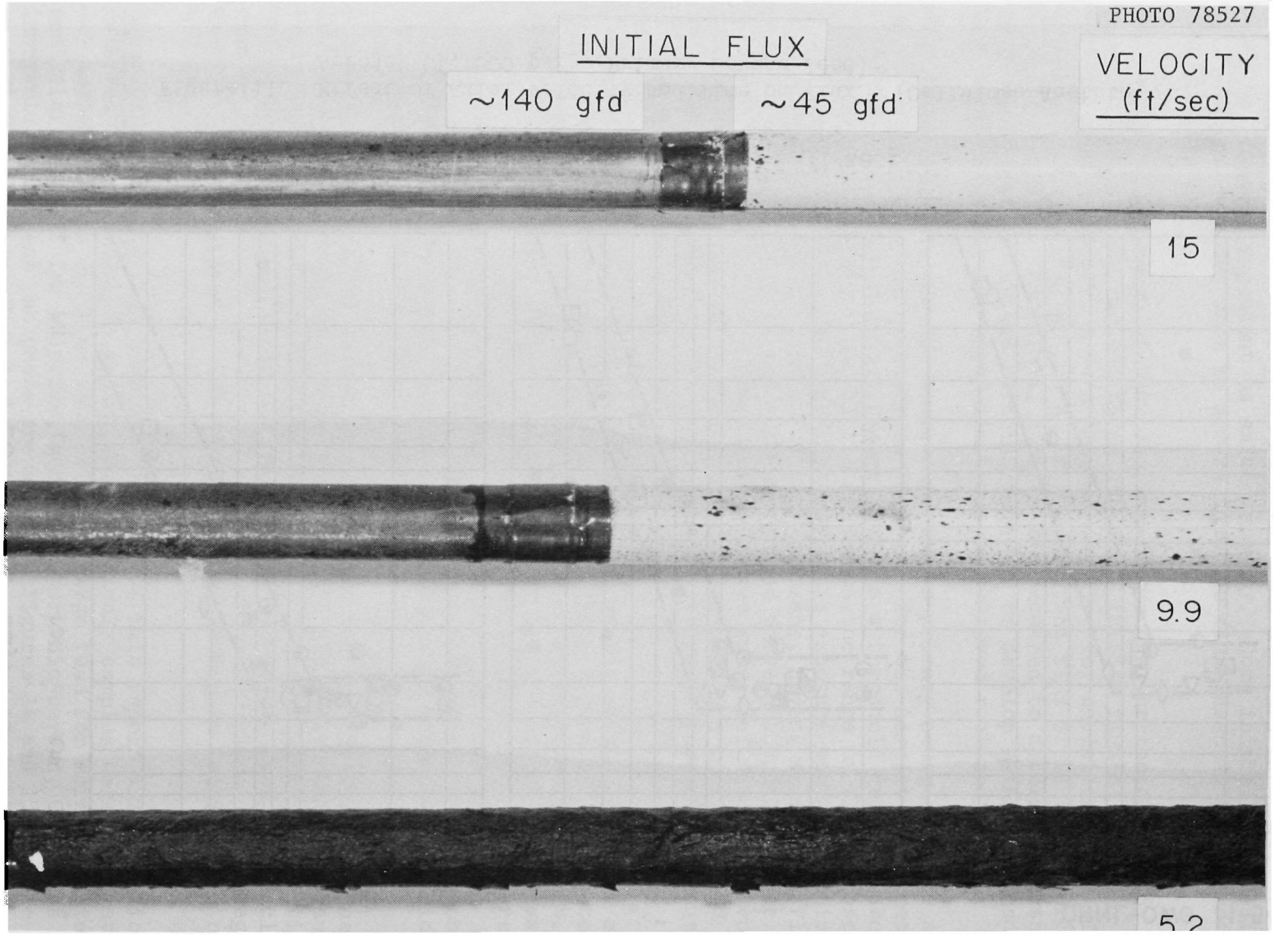

Figure 12. Membranes After 750 Hours Exposure to Primary Sewage Feed.

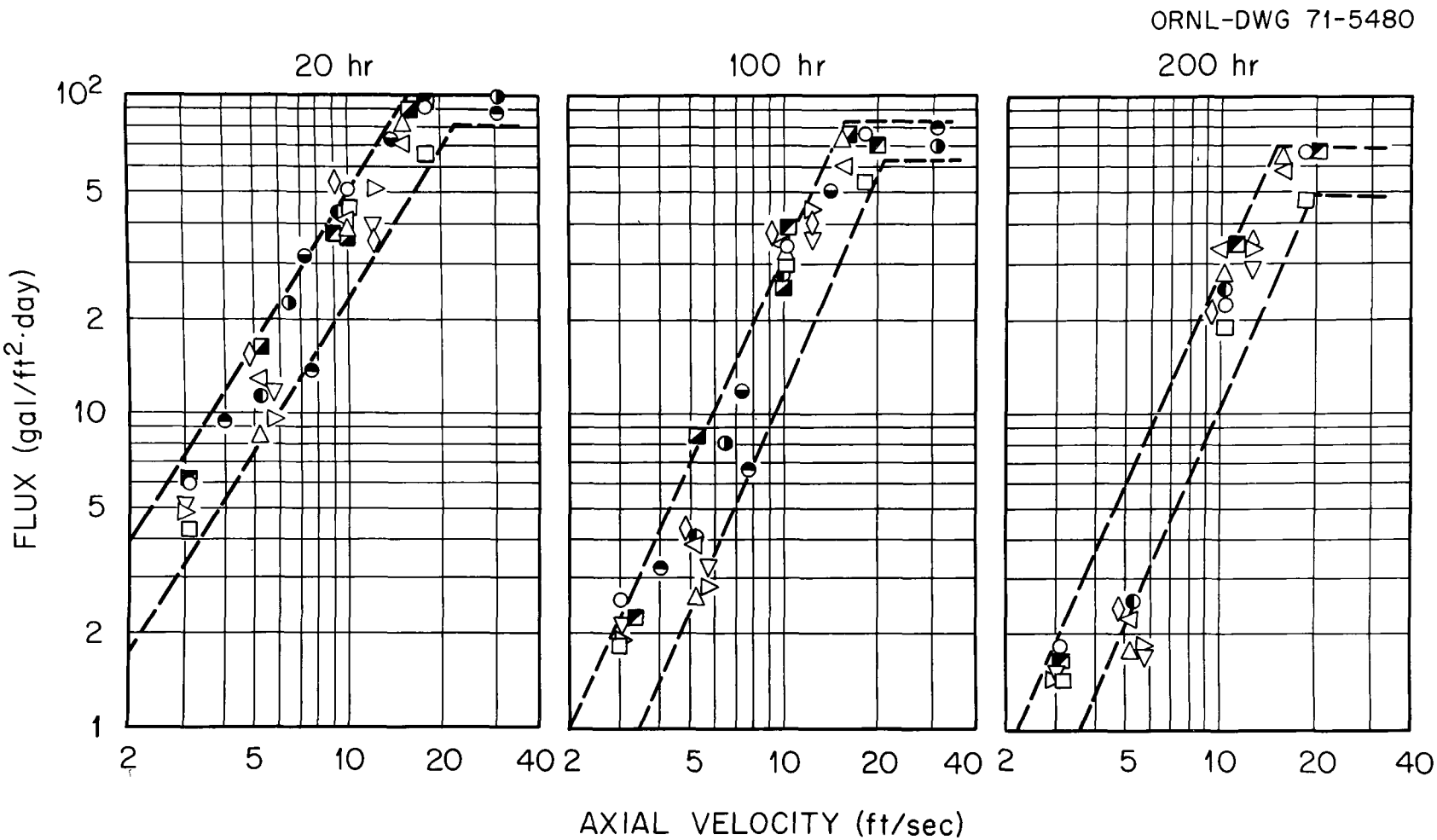

Figure 13. Effect of Axial Velocity and Time on Flux. (Cellulose Acetate, Eastman HT-00, 600 psig, Primary Sewage Feed)

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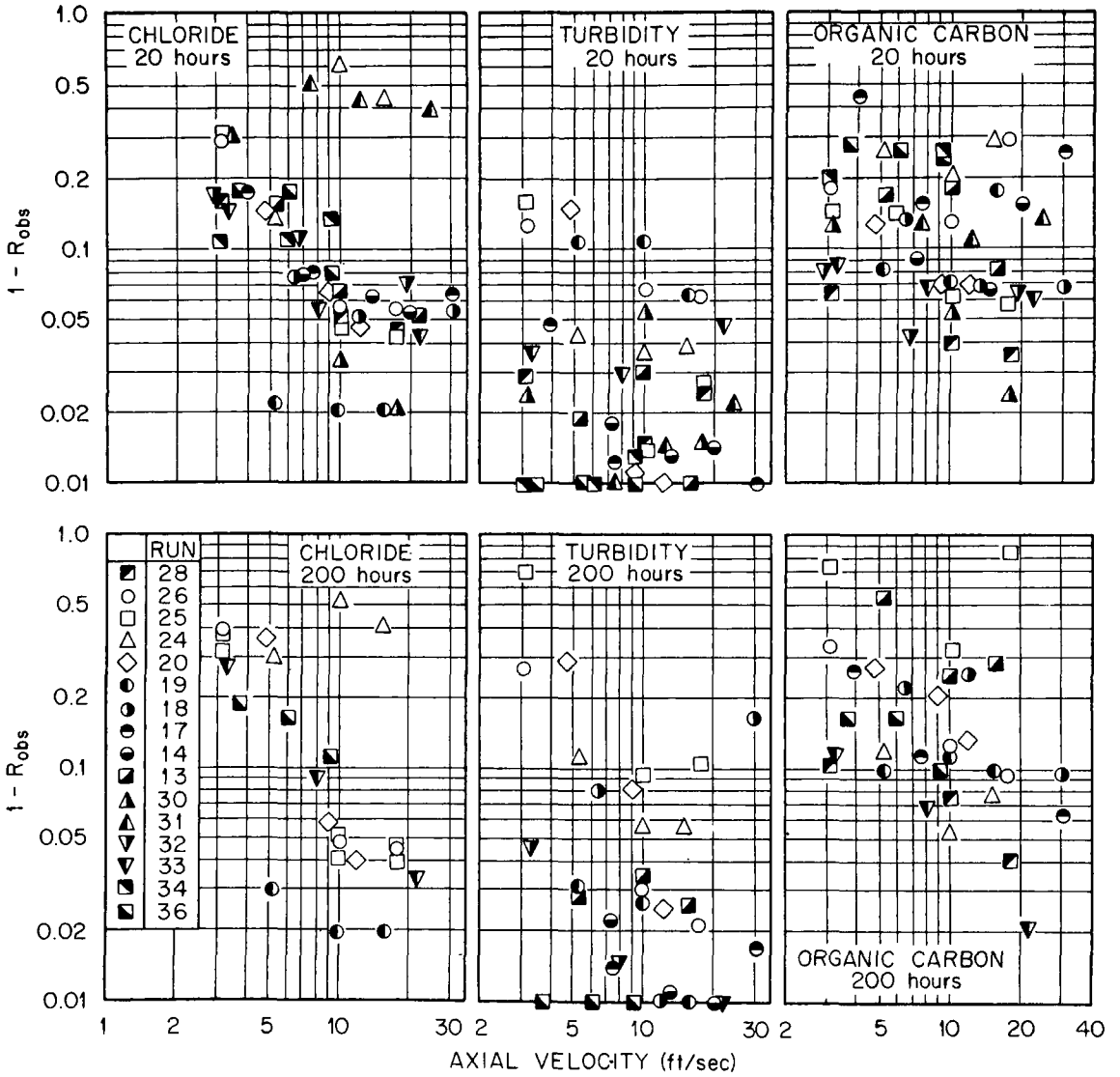

Figure 14. Observed Rejection with Aerojet 6% Cellulose Acetate Membrane with Primary Sewage Feed.

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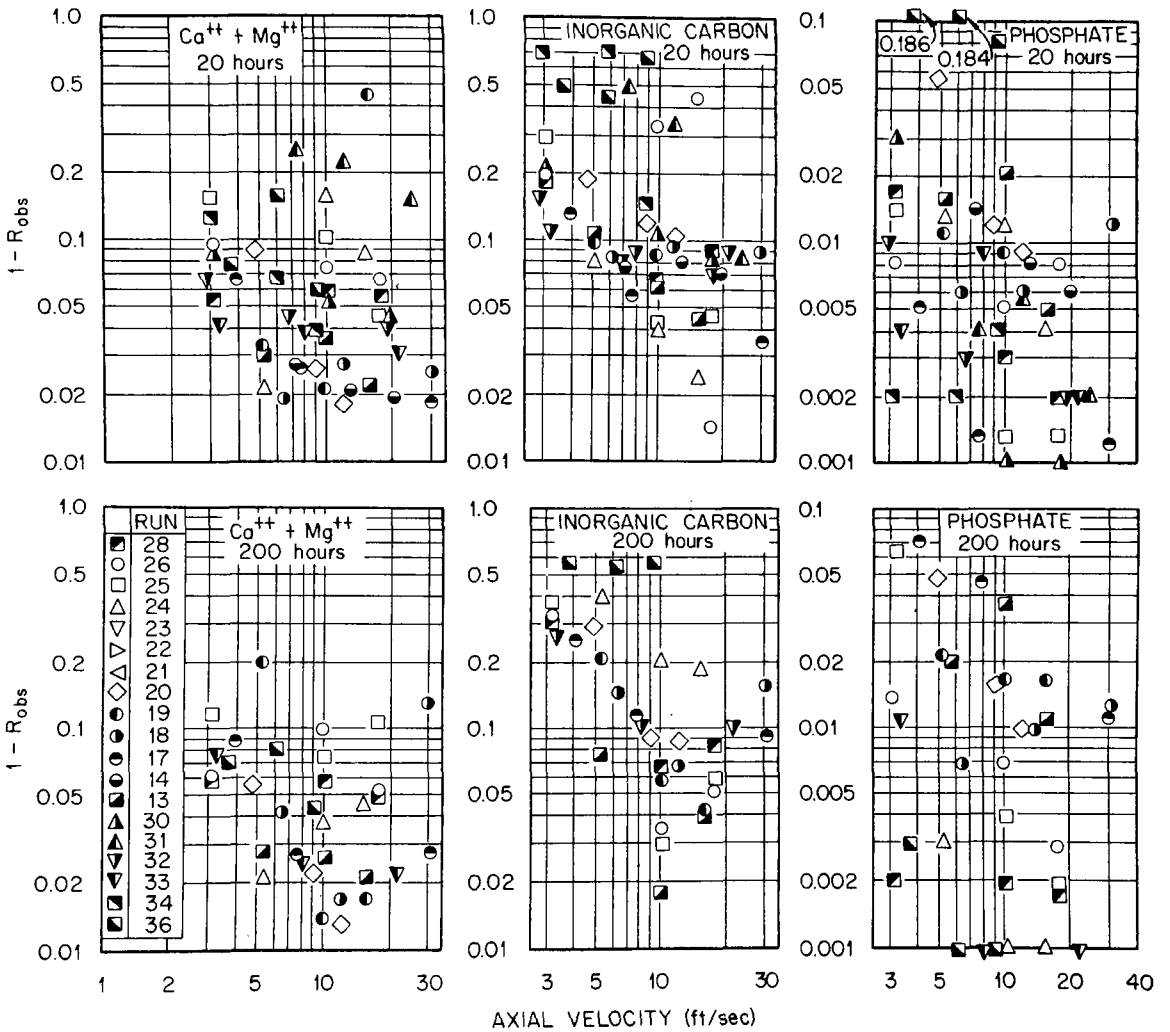

Figure 15. Observed Rejection with Aerojet 6% Cellulose Acetate Membrane with Primary Sewage Feed. (Note Scale Difference on Phosphate Rejection)

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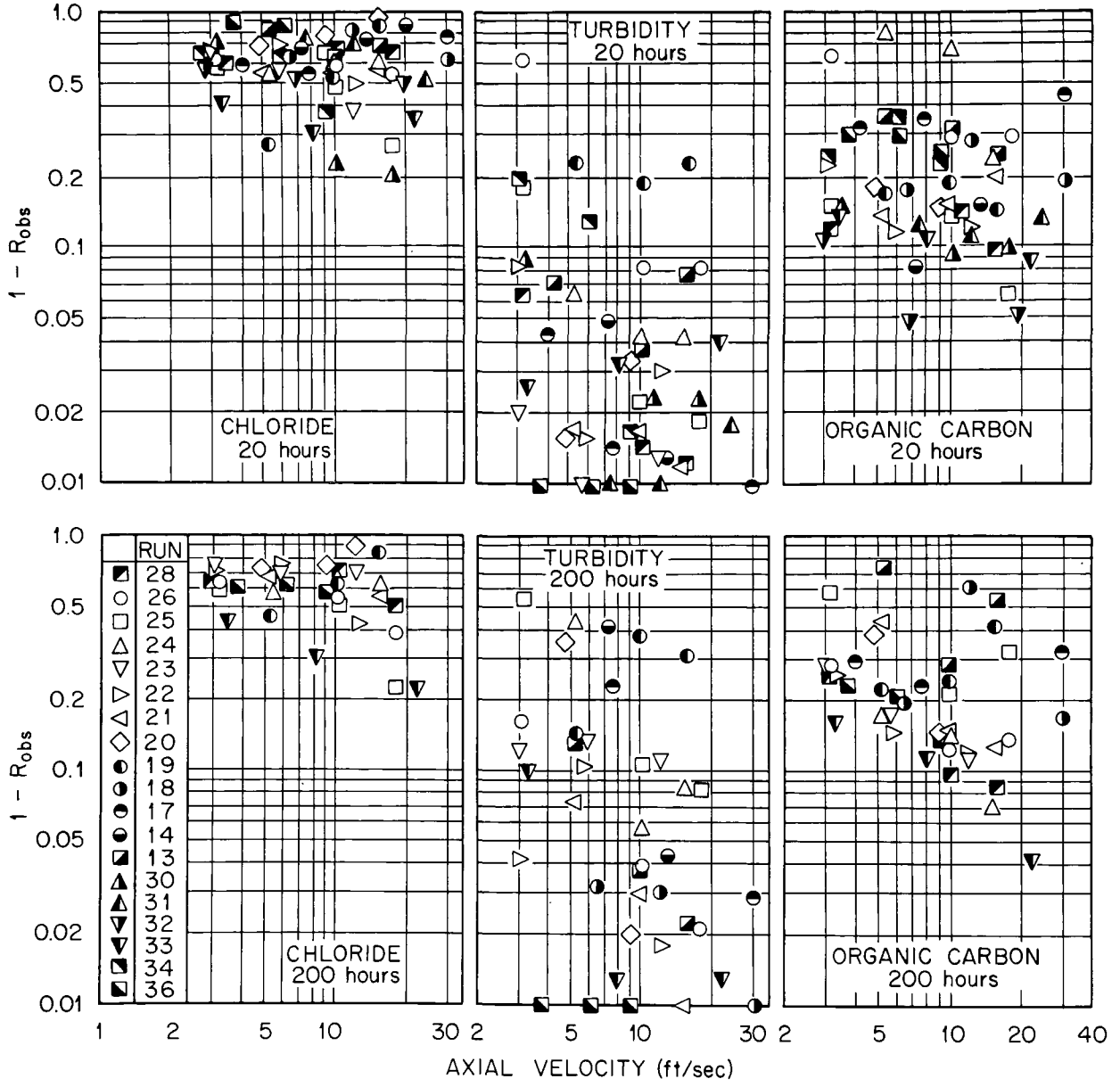

Figure 16. Observed Rejection with High Flux, Low-Salt Rejection, Eastman HT-00, Cellulose Acetate Membrane with Primary Sewage Feed.

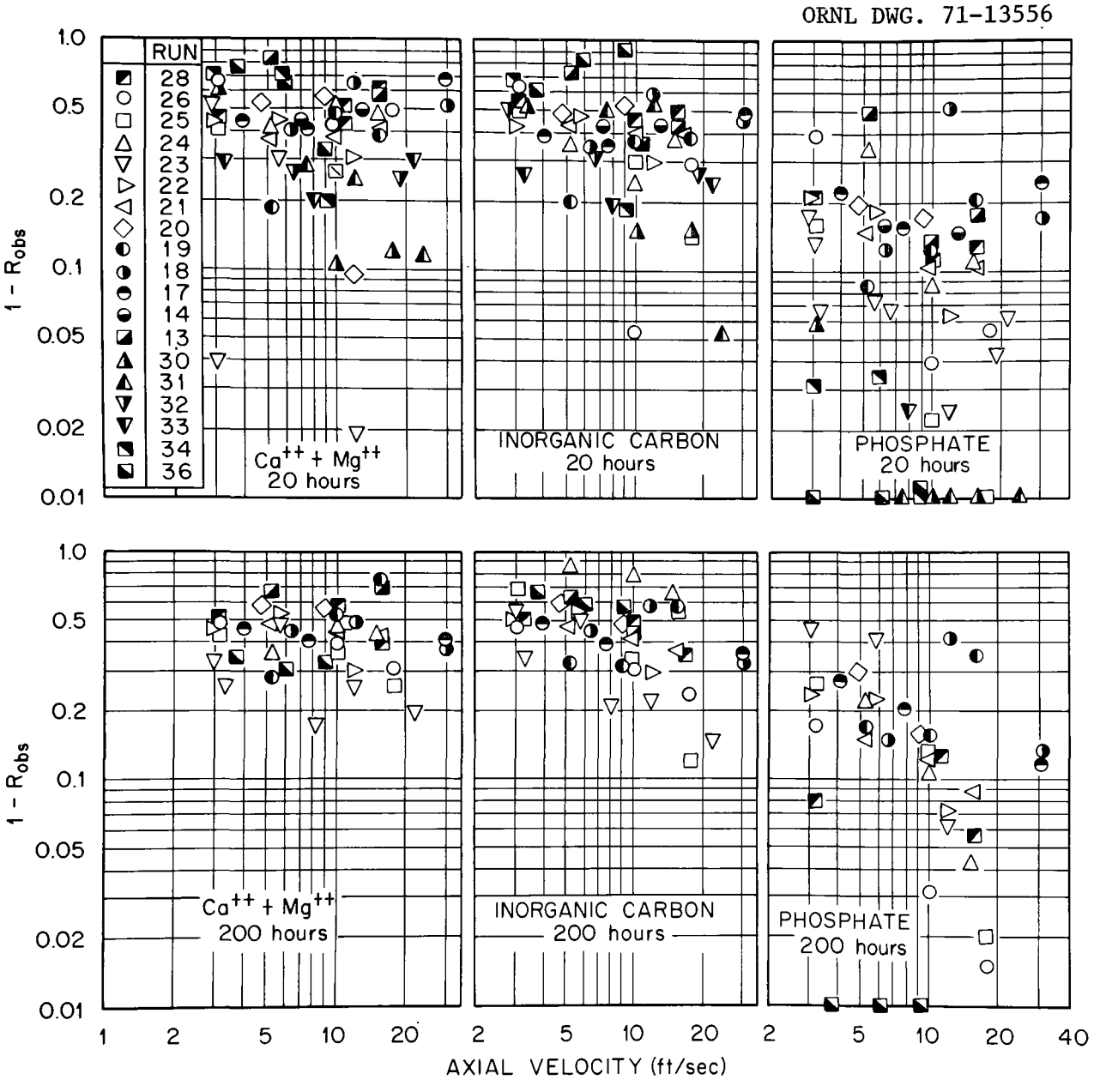

Figure 17. Observed Rejection with High Flux, Low-Salt Rejection Cellulose Acetate Membrane with Primary Sewage Feed.

zero-percent rejecting membrane. In these figures ($1 - R_{obs}$) is plotted vs axial velocity for the samples at 20 and 200 hours run time. One minus the observed rejection was chosen as the ordinate in order to spread out the high rejection results.

Although there is no clearly defined trend of rejection with velocity, there seems to be some tendency for higher rejections to be observed at higher velocities. Sheppard and Thomas (16) have shown that thick non-rejecting layers covering a membrane result in marked decrease in rejection due to increased concentration polarization in the fouling. Because thick fouling layers occurred at low velocities in the present study, a more pronounced effect of velocity on rejection was expected.

After 200 hours the median rejection and the range for the nominal 94% rejecting Aerojet 6% membrane was:

Material	Rejection, %	
	Median	Range
Chloride	94	50-98
Turbidity	97	30-99
Organic Carbon	88	15-98
Ca^{++} plus Mg^{++}	95	80-98
Inorganic Carbon	88	40-98
Phosphate	99	93-99+

These rejections compare favorably with those reported by others as shown in Table 1.

Some rejection developed with the nominally zero rejection membrane; apparently a dynamic membrane formed from sewage constituents as has been reported in previous studies. After ~200 hours the median rejection and the range for the nominal zero-percent rejecting Eastman HT-00 membrane was:

Material	Rejection, %	
	Median	Range
Chloride	40	10-75
Turbidity	90	45-99
Organic Carbon	75	25-96
Ca^{++} plus Mg^{++}	55	25-80
Inorganic Carbon	50	15-88
Phosphate	85	55-99

Although the median phosphate rejection is somewhat higher than expected, the relatively good rejection of turbidity and organic carbon is not surprising since much of this material is probably filterable and its removal does not depend on the rejection characteristics of the membrane.

Although rejection is of interest in defining membrane characteristics, in the final analysis the quality of the product water is the deciding factor. The composition of the primary sewage feed is shown as a function of time in Figure 2; run numbers are identified across the top of the

figure. Selected product analyses are shown in Figures 18 through 21. After 200 hours exposure to primary sewage, the product water compositions from the nominal 94% rejecting Aerojet 6% membrane were:

Material	Concentration, mg/ℓ	
	Maximum	Median
Organic Carbon	30	7
Ca^{++} plus Mg^{++}	30	8
Phosphate	8	0.2
Turbidity (JTU)	10	1

With the nominal zero rejecting Eastman HT-00 membrane comparable values were

Material	Concentration, mg/ℓ	
	Maximum	Median
Organic Carbon	65	12
Ca^{++} plus Mg^{++}	130	60
Phosphate	24	6
Turbidity (JTU)	10	1.5

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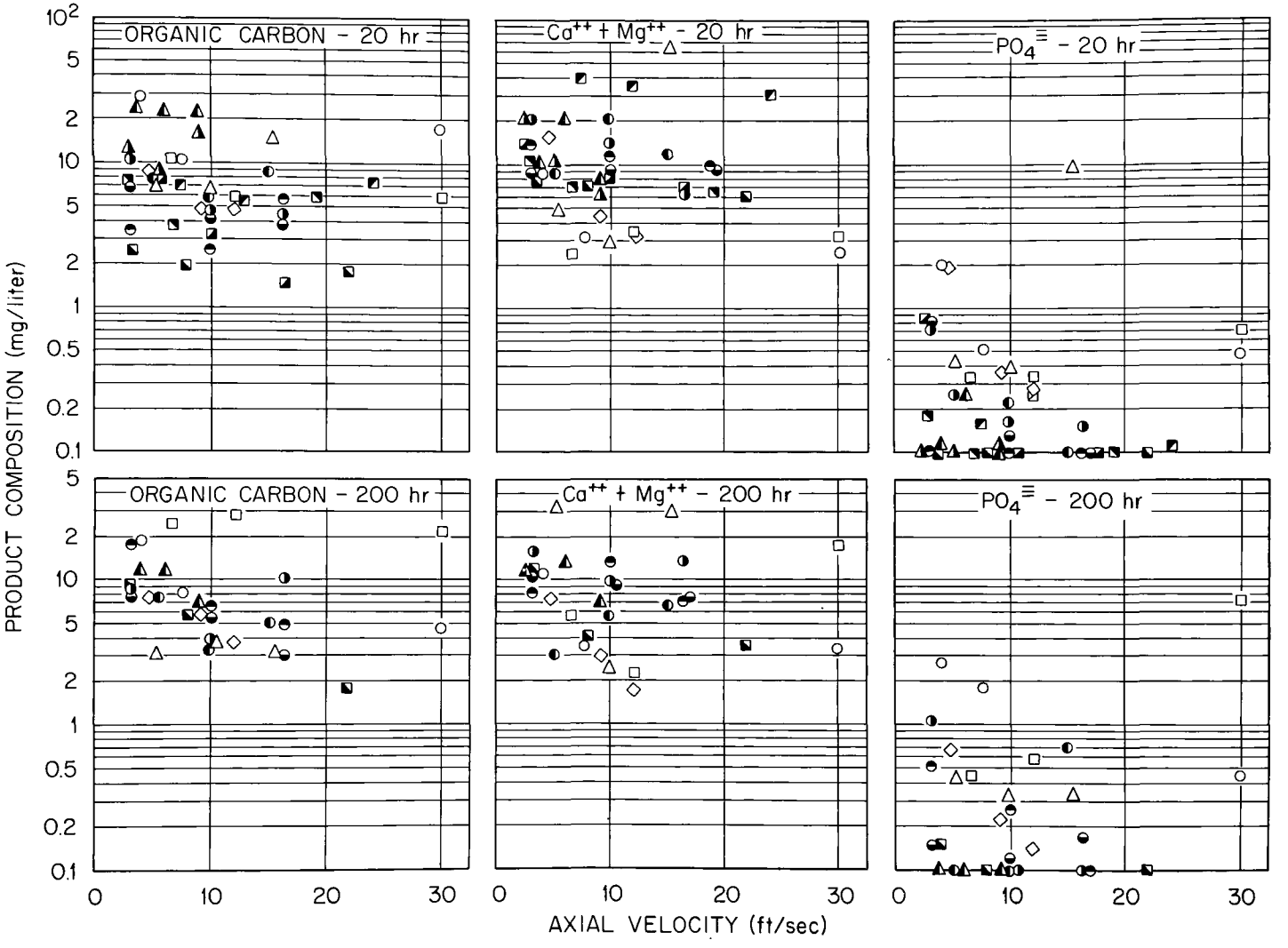

Figure 18. Product Water Composition in Hyperfiltration of Primary Sewage Effluent with a Nominal 94% Rejection Aerojet 6% Cellulose Acetate Membrane with Initial Flux of 40 to 50 gal./ft^2·day.

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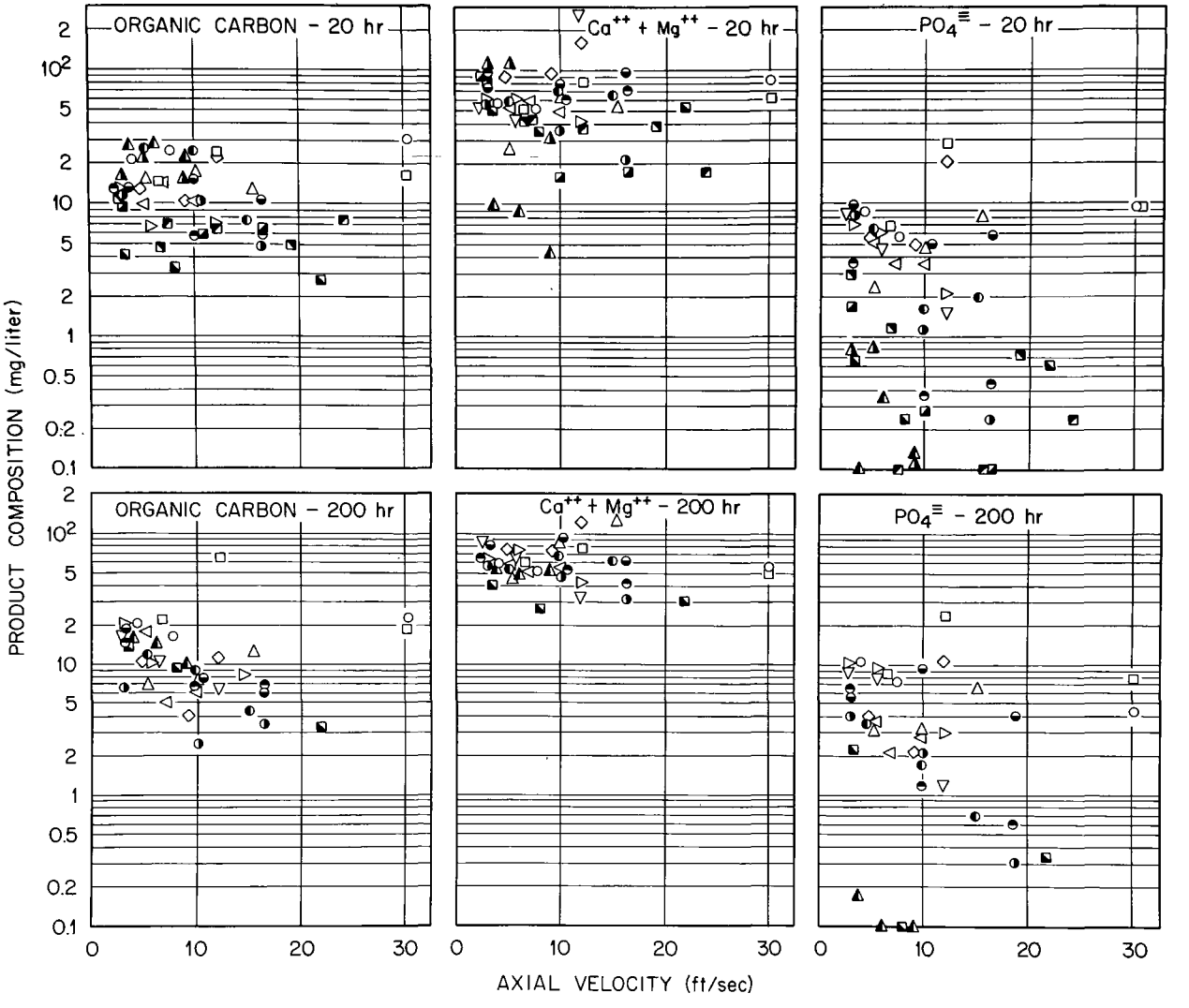

Figure 19. Product Water Composition in Hyperfiltration of Primary Sewage Effluent with a Nominal Zero-Percent Rejection Eastman HT-00 Cellulose Acetate Membrane with Initial Flux of 130 to 150 gal./ft^2·day.

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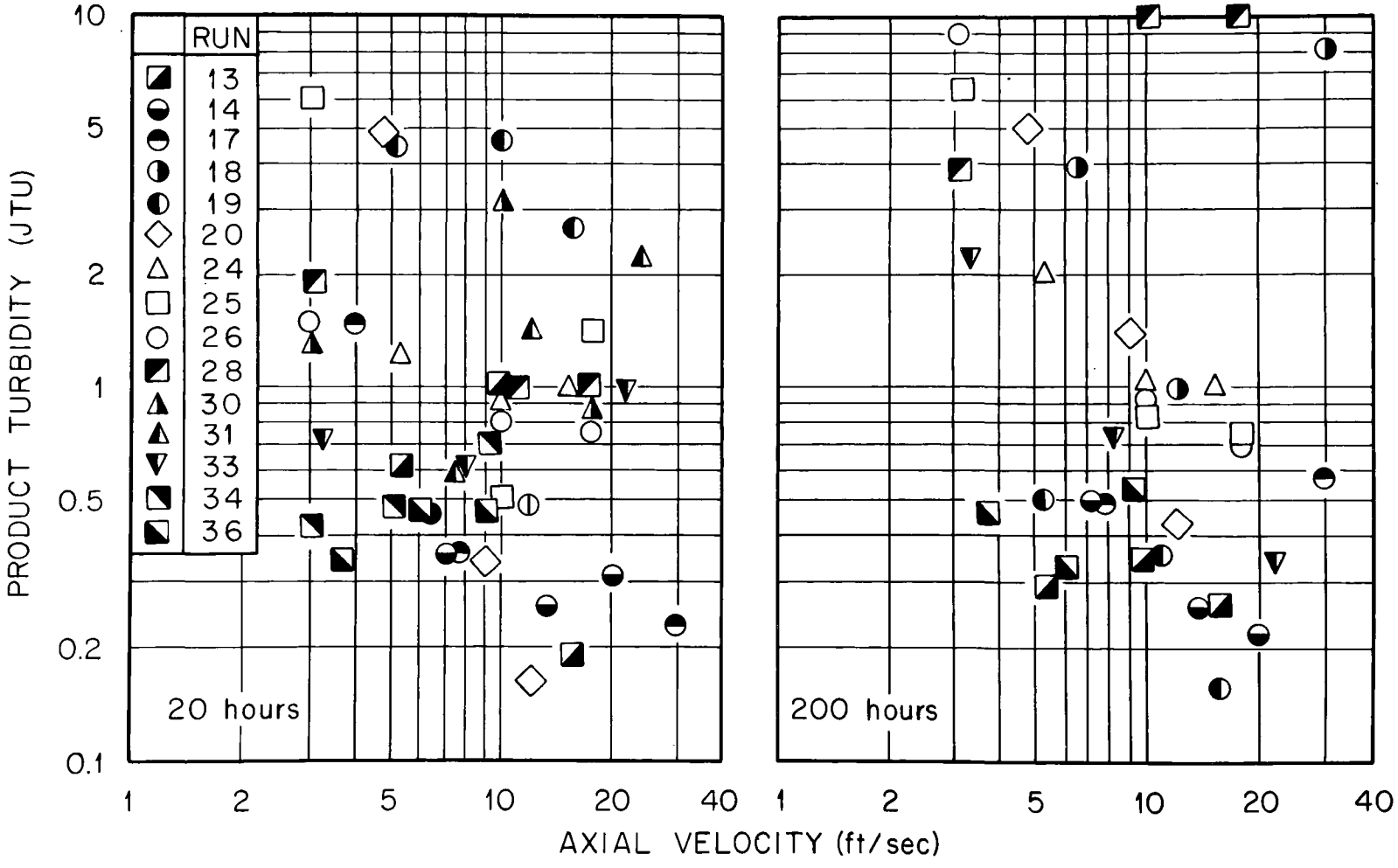

Figure 20. Product Turbidity from Nominal 94% Rejection Aerojet 6% Cellulose Acetate Membrane with Primary Sewage as Feed.

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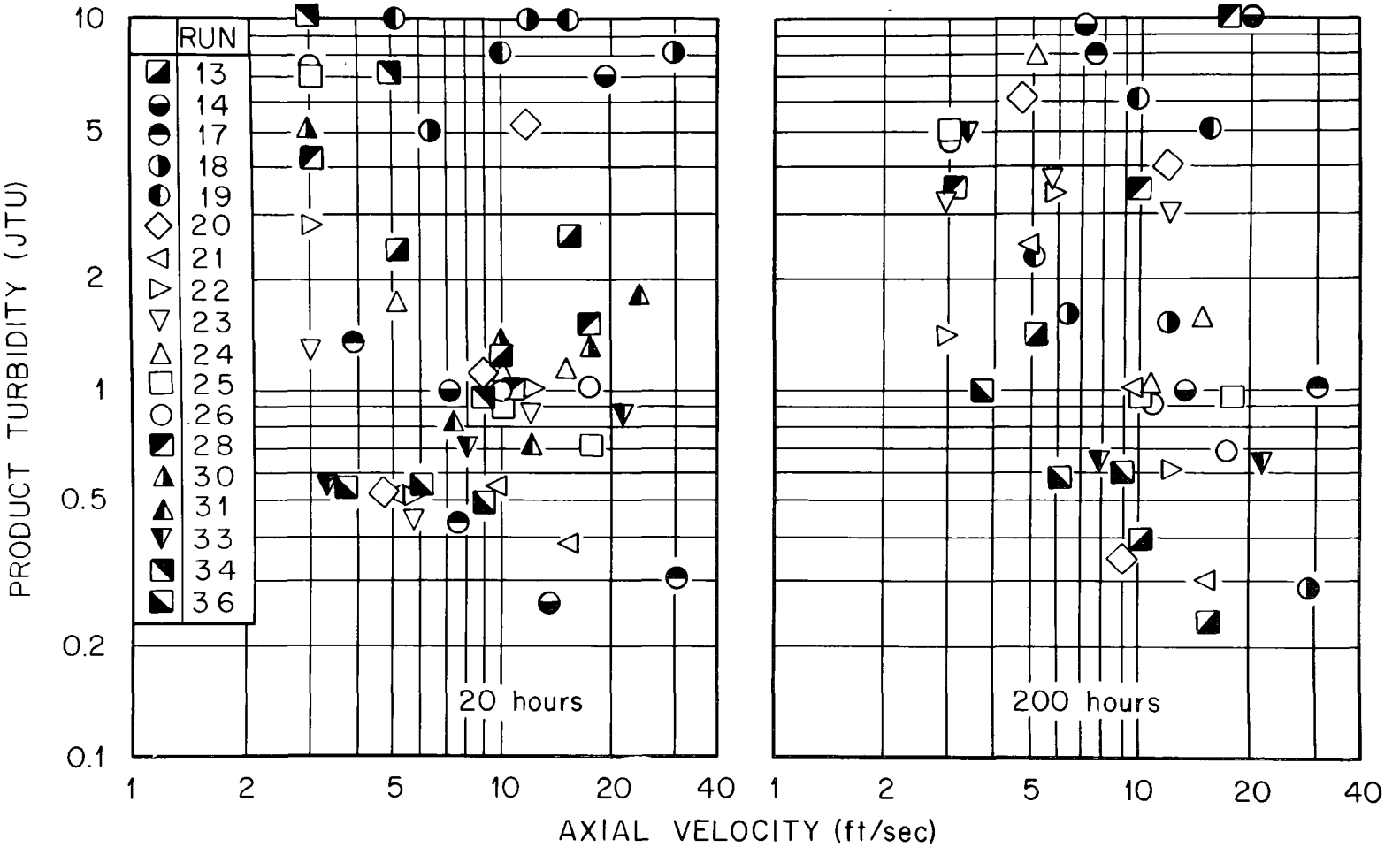

Figure 21. Product Turbidity from High Flux, Low-Salt Rejection Cellulose Acetate Membrane with Primary Sewage as Feed.

SECTION VII

CORRELATION OF RESULTS

Threshold Velocity. The results shown in Figures 9, 10, and 12 clearly demonstrate that there is a threshold velocity above which the flux decline is markedly smaller than at lower velocities. Results of fouling studies for the Office of Saline Water using untreated river water as feed are also strongly indicative of a threshold velocity.

With support of the National Science Foundation, an analysis was developed for the effect of flux on the minimum velocity required to prevent particulate solids from depositing or bouncing along the bottom of a horizontal tube. Because the limit deposit velocity given by that analysis is related to the threshold velocity for particulate fouling, the results will be briefly summarized.

The analysis was based on earlier studies of the limiting deposit (or minimum transport) velocity of flocculated particles in horizontal impermeable pipes (18, 19, 21). In those studies it was shown that Bernoulli forces accompanying the velocity gradient in the vicinity of the wall, acceleration due to the gradient of velocity fluctuations near the wall and possibly Magnus forces, all were the correct order of magnitude to play a role in preventing deposition of small particles on horizontal surfaces. Dimensional considerations suggest that, for particles with diameter smaller than the viscous sublayer and which follow Stokes' law, the friction velocity for the minimum transport condition, $(u_*)_{MT}$, may be related to properties of the particulates by

$$\frac{U_T}{(u_*)_{MT}} = K \left(\frac{D_p (u_*)_{MT}}{\nu} \right)^k , \tag{4}$$

where U_T is the terminal settling velocity of a particulate (or floc) of diameter D_p and ν is the kinematic viscosity. K and k are to be determined from theory or experiment.

If Bernoulli forces arising from the velocity gradient were assumed to be the principal lift force, then it was shown (18) that the Bernoulli forces equaled gravitational forces on the particle when

$$\frac{U_T}{U_*} = C_L \times \frac{1}{96} \left(\frac{D_p u_*}{\nu} \right)^3 . \tag{5}$$

Friction velocity may be related to the mean velocity, u, in a system by

$$u = u_* \sqrt{f/2} , \tag{6}$$

where f = Fanning friction factor.

Based on a review of existing data on lift forces the most probable value for C_L was between 0.5 and 1. Equations (4) and (5) have identical form and the exponent k has a value of 3 based on the results of the analysis.

Comparison of Equation (5) with minimum transport results obtained with both flocculated and deflocculated suspension (18,21) indicates an entirely satisfactory fit provided the left coefficient is assigned a value of 1, the upper limit of the most probable values. It seems likely that the lift coefficient in this flow regime is somewhat less than one (18). Apparently, although Bernoulli forces may play a major role in supplying particle lift in the vicinity of a wall, particle acceleration and Magnus forces also play a role and their effect is primarily reflected in the value of the coefficient in Equations (4) and (5).

When there is flow through the wall as in hyperfiltration, it is a simple matter to extend the analysis to include the additional drag force on the particle due to the product water flux, v, at the membrane surface (17). The result in the same form as Equations (4) and (5) is

$$\frac{U_T}{u_*} + \frac{v}{u_*} = \frac{C_L}{96}\left(\frac{D_p u_*}{\nu}\right)^3 \quad . \tag{7}$$

Flocculated particles in polluted waters are often of the order of 1-micron diameter. For a 1-micron particle, the terminal settling velocity is of the order 0.002 cm/min. Hence from Equation (7), any fluxes greater than 0.002 cm/min (~1 gfd) may be expected to require increased axial velocity to exceed the minimum transport condition when the particles are of order 1 micron in diameter.

Equation (7) indicates that in the limit of zero wall flux or large particle diameter (>20 microns), the velocity required to prevent particle deposition is inversely proportional to the one-fourth root of the particle diameter, while, in the limit of very small particle diameter ($D_p < 0.5$ micron), the velocity required to prevent particle deposition is proportional to the one-fourth power of the flux and inversely proportional to the three-quarters power of the particle diameter. Calculated values of the limit deposit velocity are shown schematically on Figure 22 as a function of particle diameter with the flux as a parameter. For purposes of illustration a floc density of 1.1 was assumed. Also shown on this figure is the expected variation of floc diameter with rate of shear based on results of earlier studies (20). Of course if the particles were not flocculated the diameter would be independent of the rate of shear.

Assuming that the particles in primary sewage effluent are flocculated and that the minimum transport or limit deposit velocity must be exceeded to prevent particulate fouling (and hence catastrophic flux decline) then from the results shown in Figure 4 the threshold velocity should be proportional to the square root of the flux through the membrane.

Threshold velocities determined from the 200 hour results with primary sewage feed in the present study (Figures 11 and 13) are shown in Figure 23 as a function of initial flux. Also shown on this figure are results obtained using untreated river water as feed (22), and an estimate of threshold velocity for Pomona, California secondary sewage, based on data obtained with a Havens tubular unit (7) and with a spiral wound module (3).

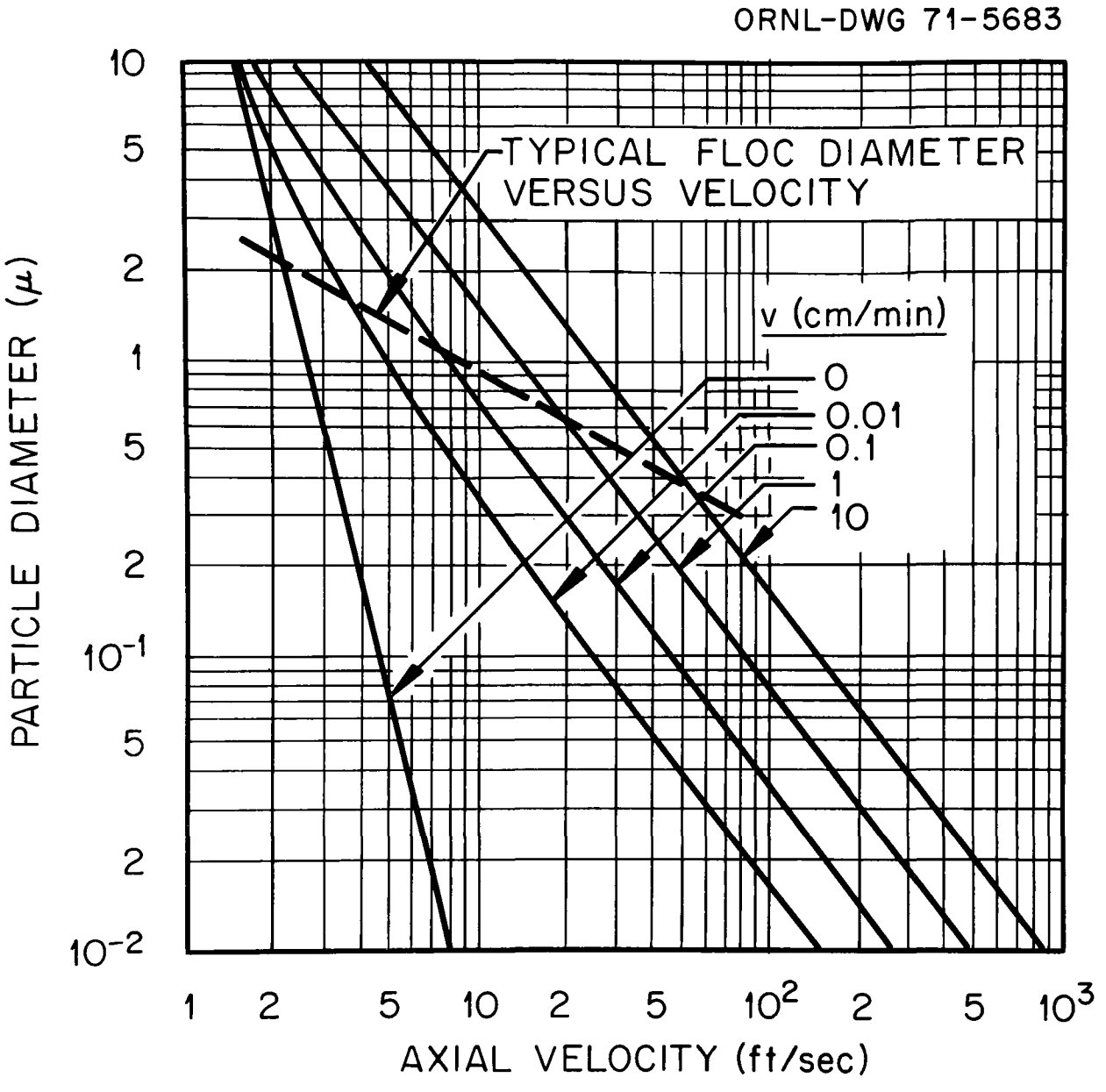

Figure 22. Effect of Particle Diameter and Flux on Limiting Deposition Velocity for Small Particles (v = 1 cm/min Corresponds to 354 gal./ft^2·day).

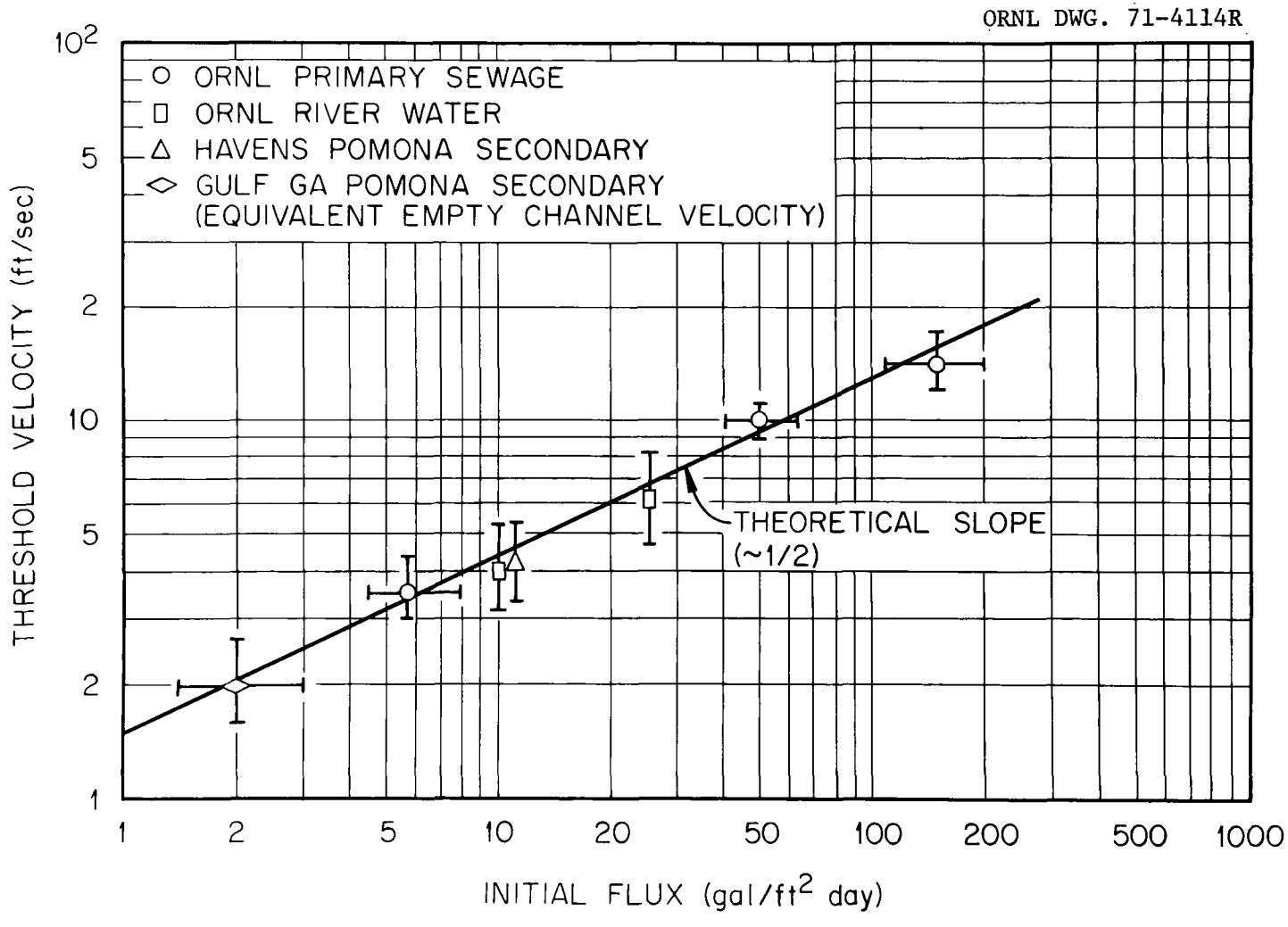

Figure 23. Threshold Velocity Above Which Catastrophic Flux Decline is Prevented with Polluted Surface Water Feeds.

In the latter case the threshold friction velocity was estimated and this was converted to an equivalent velocity for tubes using Equation (6). Surprisingly, all of the results are in rather good agreement with each other, possibly indicating similar flocculation characteristics of polluted surface waters. All results are consistent with the expectation that the threshold velocity is proportional to the square root of the initial flux. The results shown in Figure 23 may be represented by

$$u_c = (1.3 \pm 0.6)(J_I)^{1/2} \quad , \tag{8}$$

where u_c = threshold velocity in ft/sec, and

J_I = initial flux in gal./ft^2•day.

Because the "initial" flux decline at the first of a run has often been considered as due to some other phenomenon than fouling, the results for the present "initial flux declines" (Figure 10) are shown in Figure 24 as threshold velocity vs initial flux. Clearly the short time results are in good agreement with long time results. This suggests that at least the catastrophic flux declines observed in the first few hours of a test with sewage effluent, are due to particulate fouling because of poor hydrodynamics.

Both Equation (7) and Figure 22 indicate that the threshold velocity could be reduced by increasing the particle size in the feed. From Equation (7), the threshold velocity is inversely proportional to the 3/4 power of the particle diameter. Hence an increase of floc size that might be of little interest in terms of increased settling rate could result in significant reduction in threshold velocity.

As described in the section on "Additives for Sewage Floc Size Control," several different additives were tested to see whether they might be effective in increasing floc size under the high shear conditions in the loop. Although a series of experimental difficulties with the flocculants addition system prevented acquisition of conclusive results, some short time results with $FeCl_3$ as the additive indicate that this might be a promising technique. As estimate of the threshold velocity for primary sewage feed in the presence of ~50 mg/ℓ $FeCl_3$ is shown in Figure 25 as a function of initial flux. For comparison the range for untreated primary feed is shown as the cross-hatched area. Based on these scouting studies it appears that a suitable flocculant might reduce the threshold velocity to ~0.4 the value without flocculant.

Flux Decline. Although the flux decline parameter, b, ($b = -\Delta \log \text{flux}/\Delta \log \text{time}$) is of little use for extrapolation to obtain long term fluxes from short term results, it is useful as a parameter in correlating short term results. The flux decline parameter determined for times from 1 to 100 hours after primary sewage addition, is shown as a function of axial velocity in Figure 26. Three different initial fluxes are represented and the threshold velocity for each flux is indicated by the cross-hatched region.

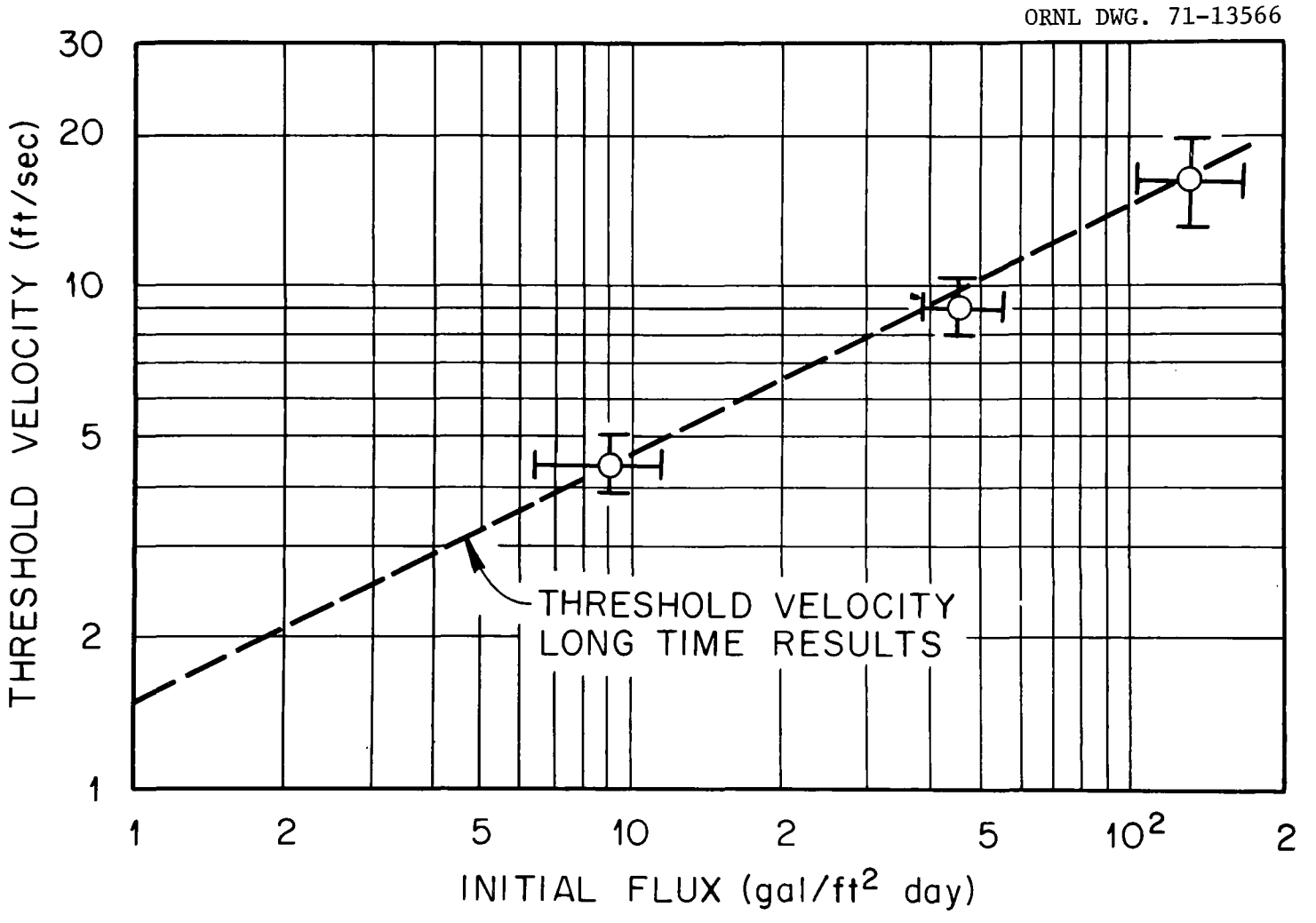

Figure 24. Comparison of Threshold Velocity from Results After 20 Hours Circulation of Primary Sewage (Round Points) with Results After 200 Hours.

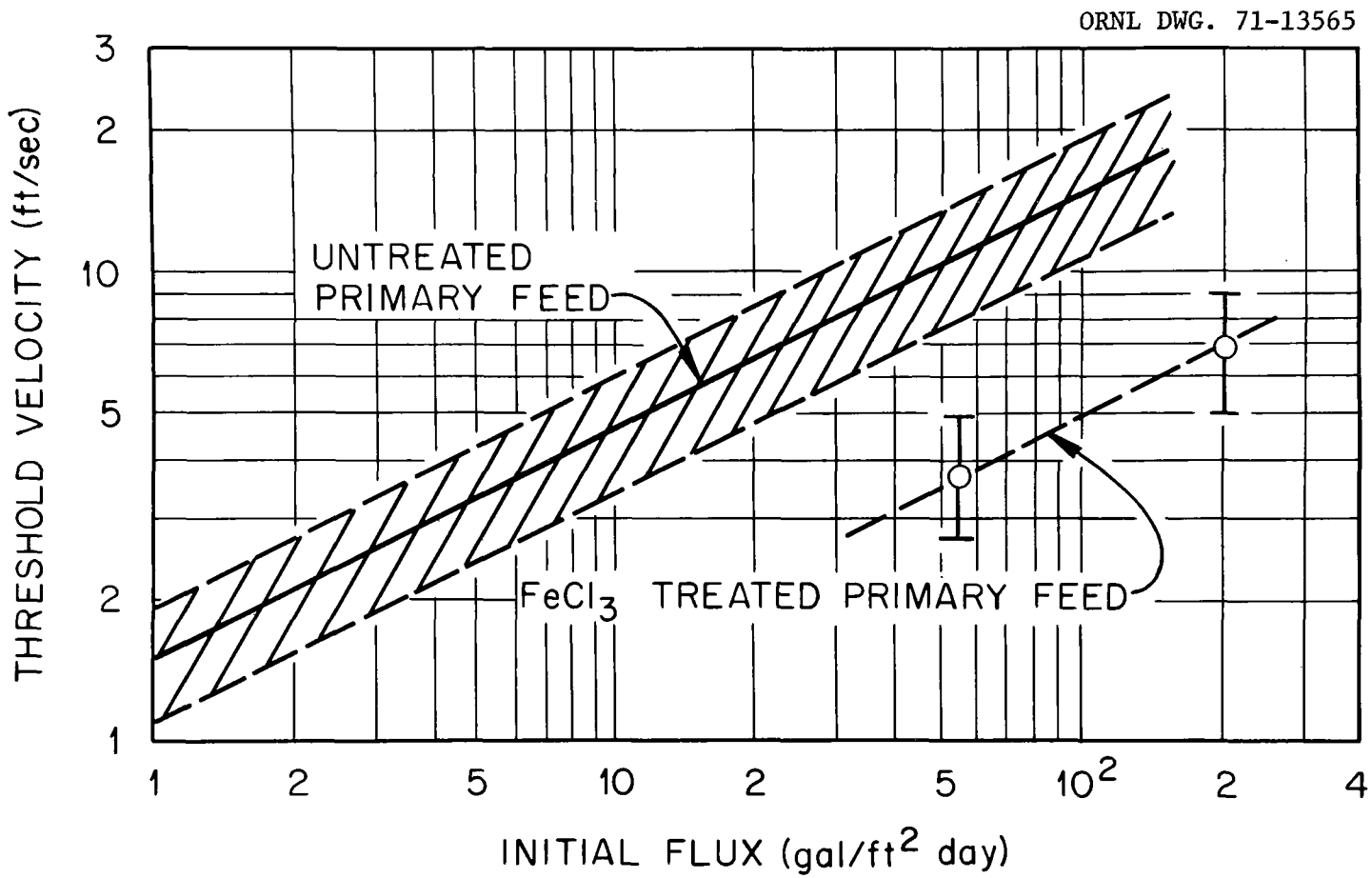

Figure 25. Effect of Adding ~50 mg/ℓ $FeCl_3$ to Primary Sewage Feed Immediately Before Introduction to Hyperfiltration Loop.

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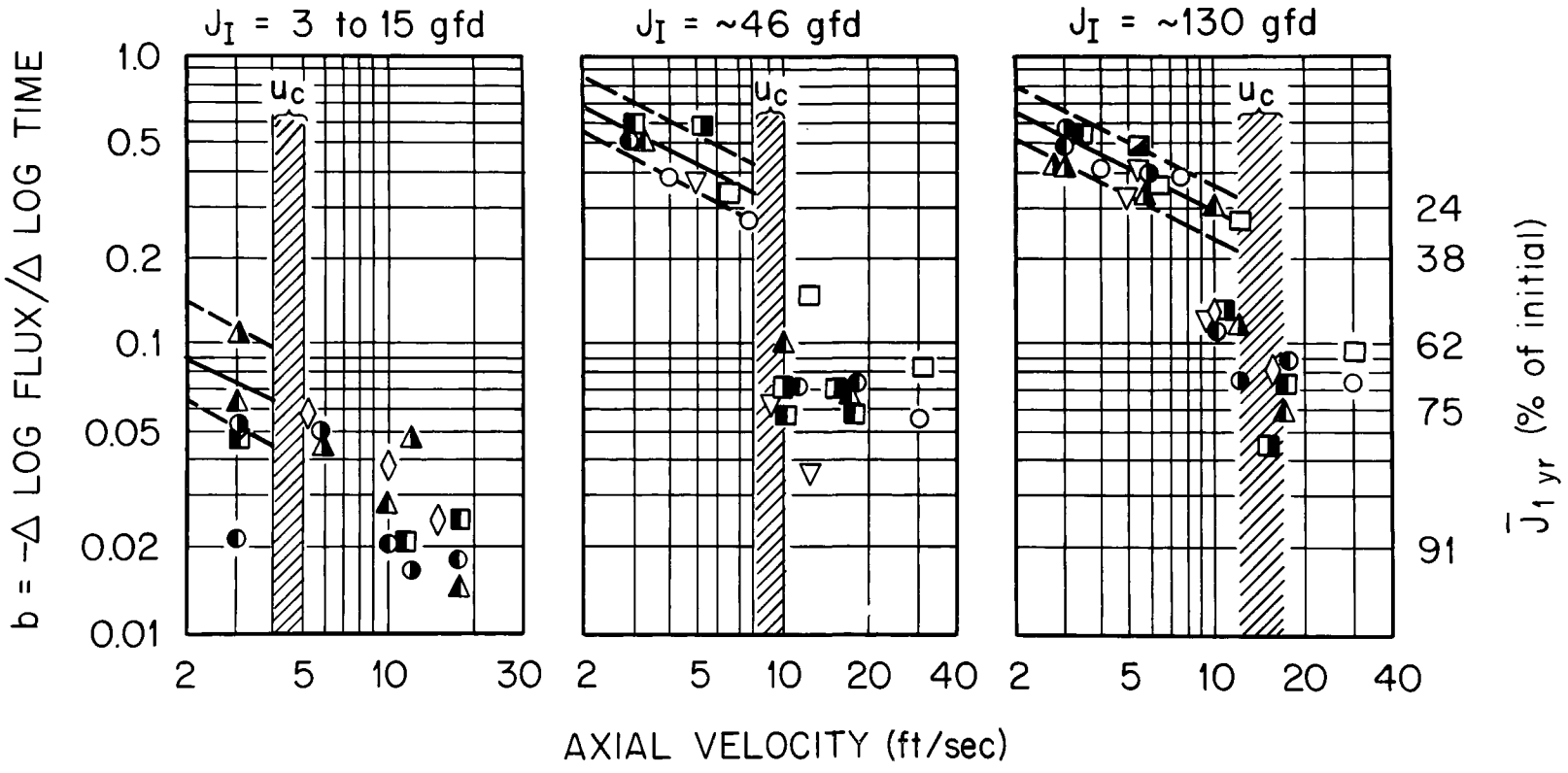

Figure 26. Effect of Axial Velocity and Initial Flux on Flux Decline Parameter, b, for Initial Portion of Runs with Primary Sewage Feed. (600 psig, Cellulose Acetate Membranes)

For the lowest initial flux, flux decline parameters were always less than 0.1, they were always less than 0.06 above the threshold velocity and b continued to decrease with increase in velocity. The median value of the flux decline parameter for velocities above the threshold was 0.025 with a range from 0.014 to 0.06.

For intermediate values of the initial flux, there was a rather pronounced discontinuity in the value of the flux decline parameter at the threshold velocity. Below the threshold velocity values of b were from 0.3 to 0.6 while above the threshold velocity the value of b had a median value of 0.065 with a range from 0.035 to 0.15.

Similar results were obtained with the highest initial flux membrane except that the median value of b for velocities above threshold was 0.09 with a range from 0.045 to 0.14.

Figure 27 is a composite plot showing values of the flux as a function of time for velocities above the threshold and times up to 700 hours. For the two higher flux membranes the points represent median values and the bars give the range of results. Because of the uncontrolled variation in initial flux with the low flux membrane, two representative runs with similar initial fluxes are shown. The flux decline parameter clearly increases with initial flux. Also the value of log flux is not a linear function of log time, with the nonlinearity increasing as the initial flux increases. In all probability this is due to increased rate of fouling by organics at the higher fluxes.

Figure 28 shows the effect of initial flux on the value of the flux decline parameter for velocities above threshold and times from 20 to 200 hours (a period when all data showed a linear log flux-log time relation). As in the previous figure the values for the two higher fluxes are indicated by median points with bars giving the range. For the lowest flux membrane, results of individual tests are given. The slope of the line drawn on the figure is 0.6 although the data are not inconsistent with values from 1/2 to 2/3.

From results studied with untreated river water it appears that the flux decline parameter is proportional to the reciprocal of the square root of the axial velocity. Although insufficient results were obtained above the threshold velocity to substantiate the velocity dependence for primary sewage feed, limited results for individual runs shown on Figure 25 are not inconsistent with a $u^{-1/2}$ dependence.

Tentatively it appears that both initial flux and axial velocity are equally important in determining flux decline and that a threshold velocity must be exceeded to prevent a catastrophic flux decline. When the velocity is above the threshold value for particulate fouling, the effect of flux and axial velocity on the flux decline parameter for times less than 200 hours is given by

$$b \propto \frac{v^{0.6}}{u^{0.5}} \quad . \qquad (9)$$

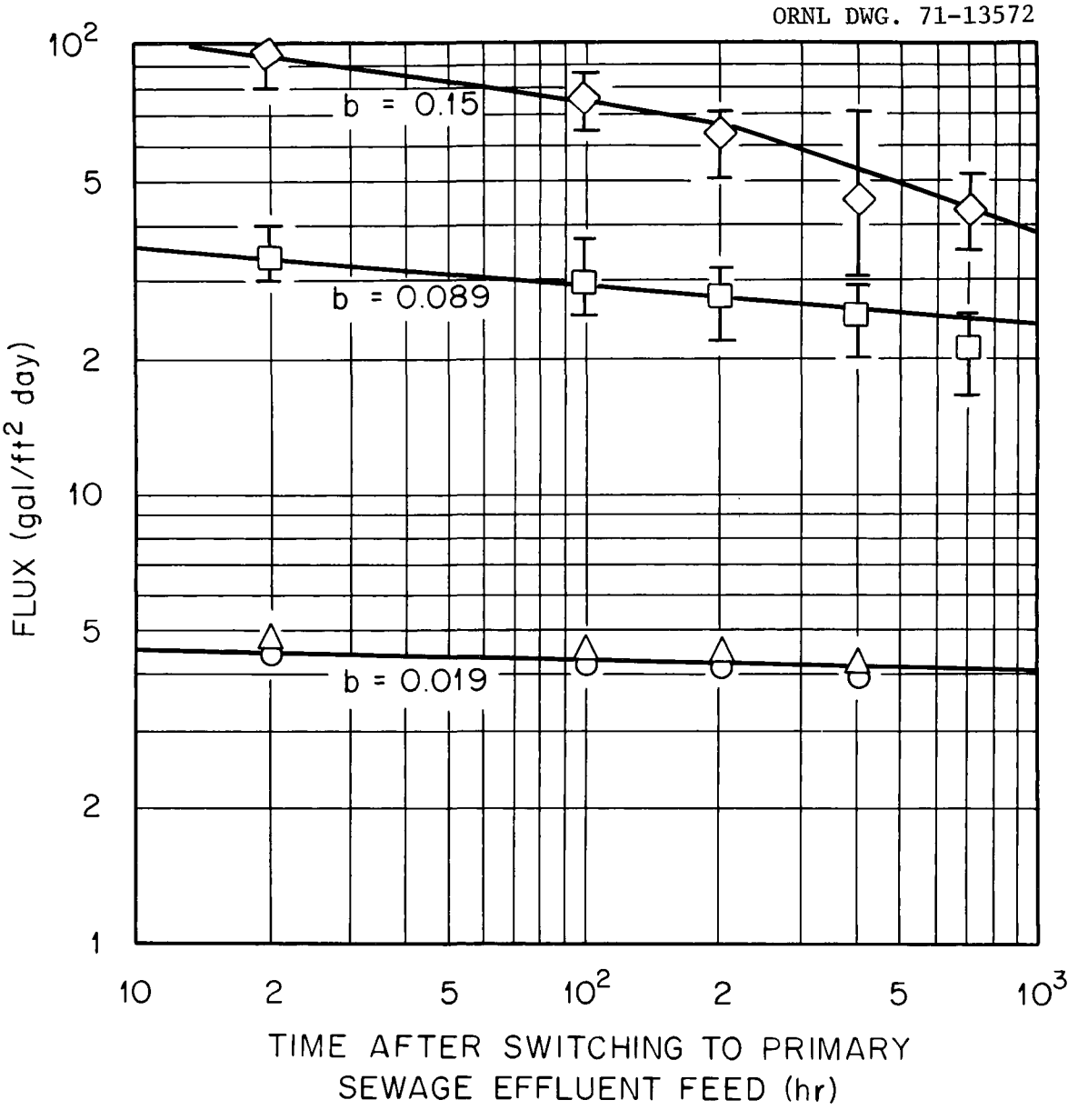

Figure 27. Summary of Flux Decline Results for Velocities Above Threshold with Primary Sewage as Feed.

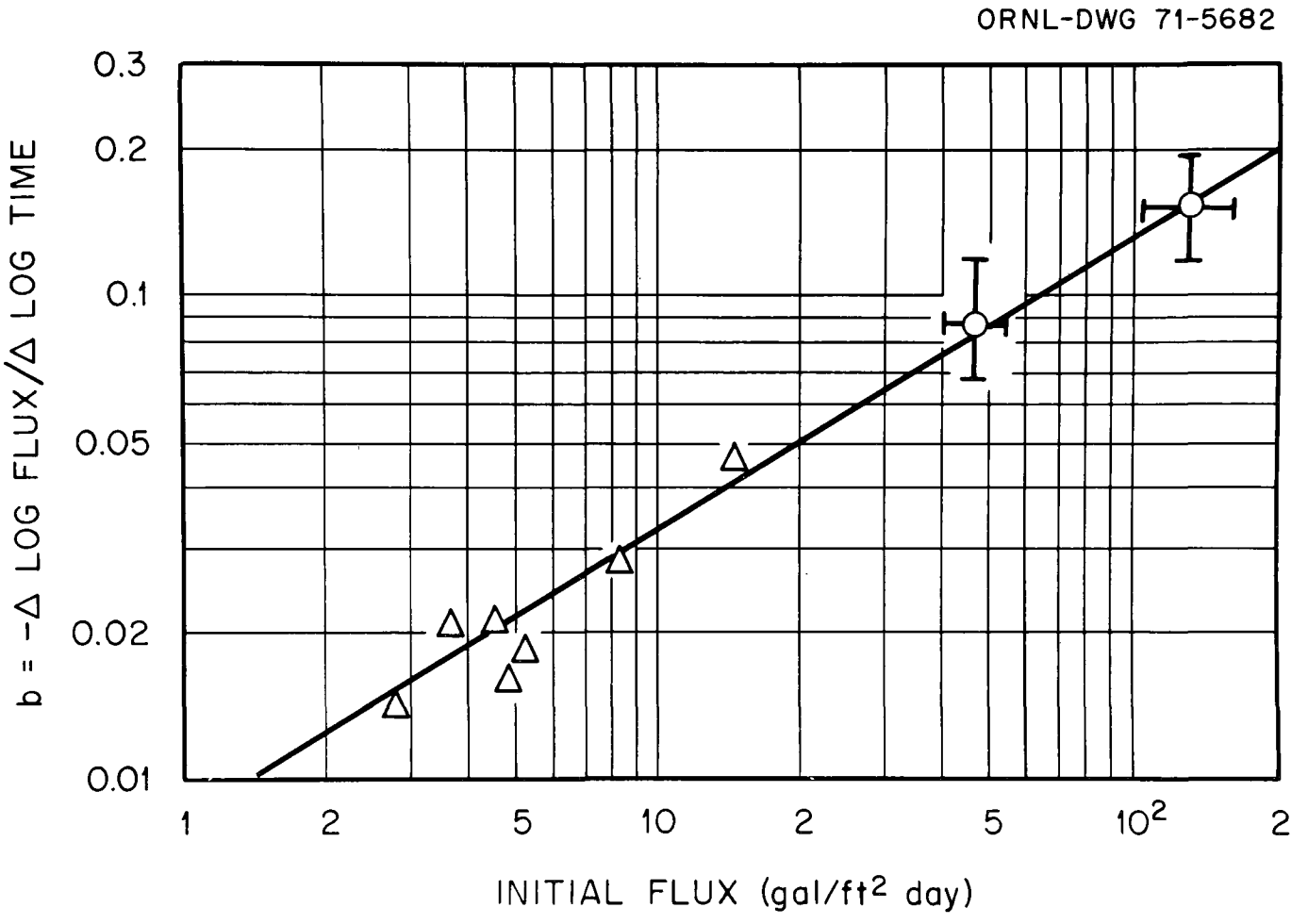

Figure 28. Effect of Initial Flux on Flux Decline Parameter, b, for Runs with Primary Sewage Feed and Velocity Above Critical (600 psig, Cellulose Acetate Membranes)

For times longer than 200 hours, the present results suggest that fouling by dissolved and colloidal organics became important. Additional studies are required to confirm this.

SECTION VIII

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SECTION IX

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16. Abstract

The effect of hydrodynamics on flux decline of cellulose acetate hyperfiltration membranes was studied using primary effluent from the Oak Ridge East Sewage Plant as feed. The system contained multiple, annular-geometry housings which could be operated simultaneously at different velocities with the same feed. The range of fluxes studied was from 2 to 150 gal./ft^2•day and axial velocities ranged from 3 to 30 ft/sec. System pressure was 600 psig.

The results demonstrate that there is a threshold velocity above which flux decline is markedly smaller than at lower velocities. Visual inspection of the membranes after a test indicates that operation above the threshold velocity markedly reduces accumulation of solids during the first 200-300 hours of a test. In scouting studies addition of a flocculant appeared to markedly reduce the threshold velocity.

During the initial 200-300 hours of those tests with primary sewage effluent as feed and in which the axial velocity was above the threshold velocity, the flux decline parameter, b, [$b = -(\Delta \log \text{flux})/(\Delta \log \text{time})$] was directly proportional to the six-tenths power of the flux and inversely proportional to the square root of the velocity.

17a. Descriptors

Particulate fouling, hydrodynamic control, threshold velocity, flux decline, hyperfiltration, reverse osmosis, waste water treatment.

17b. Identifiers

Particulates, flux control, hydrodynamics.

17c. COWRR Field & Group

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