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THE EVALUATION OF ALTERNATE METHODS OF REVERSE OSMOSIS MEMBRANE--ETC(U)
JAN 79 C A SORBER, K E LONGLEY, R F WILLIAMS DAAK70-77-C-0018

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LEVEL II

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Report No. 78-8

The Evaluation of Alternate Methods of Reverse Osmosis Membrane Maintenance

Final Report

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January 31, 1979

Supported by:

U.S. Army Mobility Equipment Research and Development Command
Fort Belvoir, Virginia 22060

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The findings in this report are not to be construed as an official Department of the Army position unless so designated by other authorized documents

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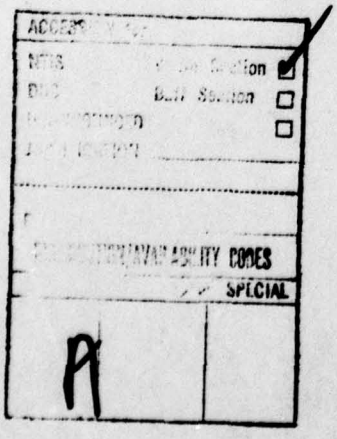

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SUMMARY

The U.S. Army Mobility Equipment Research and Development Command (USAMERADCOM) has been developing U.S. Army field water production equipment employing a reverse osmosis (RO) treatment process. During operation, material is deposited on the interior, feed-water side, of the spiral wound RO membrane. This material is capable of supporting microbiological growth which shortens the effective life of the membrane by causing a more rapid decrease in flux rate.

The purpose of this study was to determine the effects of a disinfectant on the membrane surface and to determine the bactericidal efficiencies of several disinfectants on organisms found on the membrane surface. This information may help evaluate whether field disinfection of an RI unit is practicable for prolongation of service.

Two directions of investigation were followed. The first direction studied the chemical effects of several iodine concentrations, as a suitable disinfectant, on polyamide and polycellulose special wound membranes in 2000-hour static immersion tests. The surface of the polyamide membrane was examined by scanning electron microscopy for evidence of deterioration or change. The second direction studied and compared the bactericidal efficiency of iodine, iodine bromide, and chlorine on the test organisms. The organisms employed, Pseudomonas fluorescens, Pseudomonas aerugenosa, and Escherichia coli, were isolated from a polyamide membrane subjected to

field test conditions by USAMERADCOM personnel and were determined to be the most prevalent organisms. These organisms were identified, cultured, and employed in the disinfection studies. The concentration of disinfectant was varied and the inactivation of the test organisms was measured.

Both polycellulose and polyamide membranes and their constituent parts exhibit iodine uptake with the polycellulose membrane exhibiting the most iodine demand. Static immersion studies with the polyamide membrane at three iodine concentrations under controlled conditions show considerable iodine uptake over the 2000-hour test (30 mg/1 of iodine adsorbed to a dose of 5 mg/1). Analysis of the immersed membrane pieces by scanning electron microscopy demonstrate structural changes in the membrane surface with increased immersion time for all iodine doses employed (1, 5 and 50 mg/1). The appearance of holes or voids in the membrane surface is observed upon immersion in the iodine solutions but not on samples immersed in the buffer medium alone.

Disinfection studies for the test organisms using iodine, iodine biomide, and chlorine demonstrate that very effective bactericidal inactivation can be obtained for all the disinfectants at the higher concentrations levels. At lower concentration levels, one to two logs bacterial kill is observed for all disinfectants.

The major conclusions derived from this study are:

1. P. aeruginosa, P. fluorescens, and E. coli are the predominant microbiological agents on fouled polyamide membranes (Potomac River water test site) and these organisms can be isolated, identified, and cultured.

2. There is significant polyamide membrane deterioration upon immersion of the membrane in a static test system. The deterioration is evidenced by the appearance of holes of various sizes in the surface. These structural abnormalities appear after 45 days immersion in either a 1 or 5 mg/l iodine solution.

3. The organisms P. aeruginosa and P. fluorescens are more resistant than E. coli to inactivation using either iodine, iodine bromide, or chlorine as the disinfectant.

4. Iodine and iodine bromide are excellent bactericidal agents with efficiencies that are comparable to the chlorine disinfection system used in this study.

PREFACE

Authorization for the work described in this final report is according to Contract No. DAAK70-77-C-0018 with the U.S. Army Mobility Equipment Research and Development Command, Fort Belvoir, Virginia 22060.

INTRODUCTION

Background. The U.S. Army Mobility Equipment Research and Development Command (USAMERADCOM) has carried out research for the development and application of cellulose acetate and polyamide membranes for use in a new generation of U.S. Army field water production equipment employing the reverse osmosis (RO) process. During operation dissolved, colloidal, and some suspended material is deposited on the interior, feed-water side of the spiral wound membrane as the permeate passes through to its exterior. This material contains nutrients which support microbiological growth on the interior of the membrane which, in turn, shortens the membrane life by causing a more rapid decrease in flux rate. The microbiological growth might be controlled by either dosing with high concentrations of chlorine or by maintaining a chlorine residual at the interior face of the membrane. However, chlorine, being a strong oxidant, readily hydrolyzes the membrane material. This action of the chlorine results in an unacceptably rapid membrane deterioration. Thus, a microbiocidal or microbiostatic chemical must be identified for addition to the feed-water which possesses the following ideal characteristics:

1. Prevents proliferation of microbial growth on the interior surface of a membrane.

2. Does not cause deterioration of membrane or materials used in element construction.

3. Is relatively safe and easy to inject into the feed water stream by operators.

4. Appears in the permeate as an effective disinfectant, but is not toxic to man in expected concentrations either as the chemical initially applied or as a by-product of the original chemical form.

5. Forms a residual which is easily monitored under field conditions with existing technology.

6. Is economical.

Membranes. The dry-RO polyamide membrane employed in this study is comprised of three regions which includes a thin, dense skin and a substructural region which provides support for the skin and which has a much lower density due to the presence of open cell voids. In some cases an intermediate zone of closed cell voids exists between the skin and the substructural region. This zone has a density between that of the skin and the substructural region. The substructural voids provide for rigidity of the membranes, and due to their large size, dry-RO membranes will not collapse upon drying (1). Structural changes in the membrane should not occur upon drying of the polyamide membranes. On the other hand wet-RO membranes (polycellulose acetate) are composed of a small void size which results in a substantial internal surface area. Upon drying of a wet membrane, the capillary forces, which are exerted, would lead to structural changes; void collapse and membrane densification occurs, causing a precipitous decline in permeability (1).

Kesting (1) reported the presence of spherical-like structures on the surface of dry-RO cellulose acetate membranes. The hypothesis that these structures might be artifacts was rejected based upon the available information. Kesting (1) further characterized the spherical-like structures as nodules which appear to be slightly ellipsoidal with a long axis of 200 $\mathring{A}$. The origin of these nodules is not known, however, they appear to be characteristic of dry-RO cellulose acetate membranes.

Disinfectants. Among the disinfectants considered for this study were iodine and iodine bromide. Both iodine and iodine bromide are solids at normal conditions of temperature and pressure; however, iodine bromide sublimes rapidly slightly above room temperature. Nevertheless, their storage and application characteristics might be more favorable under field conditions than a disinfectant such as bromine which is in a liquid state at normal conditions of pressure and temperature. Iodine and iodine bromide have a marked advantage since iodoamines are not produced in detectable concentrations. Therefore, halamine formation does not adversely affect their microbiocidal properties as observed with a disinfectant like chlorine. The microbiocidal properties of iodine have been well documented (2, 3, 4, 5, 6). While iodine demonstrates good microbiocidal properties, its efficiency, on an equimolar basis, is reported to be less than that of chlorine (7, 8). As a weaker oxidant, relative to the other halogens, it might have negligible reaction with membrane materials.

Both ozone and chlorine dioxide, while being good microbiocides, are both very strong oxidants, and share the common disadvantage of requiring manufacture on-site. Furthermore, the ozone residual does not persist.

Ultraviolet light is an effective disinfectant but it does not result in a residual, nor can it readily be applied to the interior of a spiral wound membrane. The two interhalogens, bromine chloride and iodine monochloride, were excluded from consideration due to their relatively high reactivity. Bromine is a very corrosive and toxic liquid which makes its use in the field difficult and hazardous, eliminating it as a candidate disinfectant.

While the active microbiocidal agent of the chlorinated isocyanurates is hypochlorous acid, the slow chlorine release of these compounds may permit the use of chlorine concentrations sufficiently low to effect insignificant

membrane deterioration, yet provide acceptable microbiocidal or microbiostatic properties. The toxicological properties of these compounds have not been assessed at expected concentrations in the finished water.

The chloramines and bromamines (2, 6, 9, 10, 11) have demonstrated microbiocidal properties but are far less efficient as oxidants relative to their parent halogen. Therefore, future studies could include chloramines as a possible candidate disinfectant. The bromamines initially were included as a candidate disinfectant in this study, but subsequently were excluded from the study due to their instability as discussed elsewhere in this report.

The candidate disinfectants selected for study were iodine, iodine bromide, and chlorine. Disinfection data for chlorine was primarily for comparative purposes, since considerable knowledge exists concerning its microbiocidal properties. The same test geometry as iodine and iodine bromide was used for chlorine in order to obtain baseline data upon which to judge the performance of the latter two candidate disinfectants.

OBJECTIVES

The two major objectives of this study were:

1. Determination of effect of iodine on membrane material.
2. Determination of the relative bactericidal efficiency of iodine, iodine bromide and chlorine.

EXPERIMENTAL

Membrane Immersion. Virgin spiral wound polyamide and polycellulose membranes, furnished by USAMERADCOM, were used in this phase of the study to evaluate the effects of iodine on the membrane material.

Experiments designed to obtain preliminary information regarding the behavior of the target membranes and candidate disinfectants, as well as protocols for analysis and experimental validation were performed upon dry, cut (1-2 by 4-6 cm) membrane pieces. The backing, spacer, and membrane itself were physically separated and their responses were measured individually. Sections of the spiral wound membranes used for analysis were chosen by a visual inspection for uniformity of the surface that was free from holes or blemishes.

Initially, membranes were to be immersed in unbuffered iodine solutions in covered Nalgene tanks. However, control experiments demonstrated that the iodine residual loss averaged approximately 0.8 mg/l/day due to reaction of the iodine with the walls and bottom of the tank. Also, a minor loss resulted from the volatilization of the iodine. Subsequent investigations using glass chromatography tanks with glass covers have yielded an iodine loss rate of approximately 0.0033 mg/l/day providing that a constant head space in the tank was maintained. When the head space in the tank was increased, iodine loss rates up to 0.17 mg/l/day were observed. Therefore, glass chromatography tanks with 4.4 l capacity were treated for chlorine demand and washed with demand free water. The subsequent iodine immersion studies were performed with a constant head space of 0.25 in. from the top of the tank.

Proper control of pH of the immersion solution was essential for complete system description. Since a phosphate buffer of 0.1M (pH=7.00) produced no

visible changes upon either the polyamide or polycellulose acetate membranes in week long trials, a phosphate buffer was chosen for the immersion studies. A phosphate buffer concentration of 0.01M (ten-fold lower than most concentrated control experiments) was employed in the 2000-hour immersion study to minimize any buffer effects. The pH was monitored at each solution change and was constant to 0.1 pH units over the entire study period (pH=6.9-7.0). A control buffer solution without iodine was also examined with membrane present for the 2000-hour study period.

All stock solutions of iodine in buffer were allowed to stand for at least two hours prior to concentration determination by amperometric titration due to an iodine demand of arsenate in the phosphate buffer.

Demand-free water for all solutions was prepared by passage through an anion-exchange resin, glass distillation, and final passage through an activated carbon column.

Buffered solutions containing 0, 1, 5, and 50 mg/l of iodine were introduced into the covered glass chromatography tanks. Sample pieces of the polyamide membrane (7.62 cm by 17.78 cm) were introduced into the four chromatography tanks. Concentration excursion of iodine was to be kept less than ±10 per cent requiring 200 ml samples to be withdrawn from the tanks and analyzed by amperometric titration at frequent intervals. During the 2000-hour study, adjustment of the iodine concentration in each tank was made by changing the entire solution. The new solutions had an iodine concentration identical to the initial solution. Each tank was protected from light by an aluminum foil cover. At each examination of iodine concentration the pH of the solution in each tank was monitored. Temperature was ambient (26C) and invariant. At intervals of 0, 0.25, 0.5, 1, 2, 3, 5, 10, 30, 45, 60, and 83.3 days 0.64 cm by 7.62 cm pieces were removed from each membrane in the

four tanks. These membrane strips were stored wet in sealed glass vials and protected from light for future preparation for SEM analysis.

Scanning Electron Microscopic (SEM) Analysis. Polyamide membrane pieces of approximately 0.5 cm by 0.5 cm were cut by a scapel from the 0.64 cm by 7.62 cm strip removed from the large membrane piece which had been immersed in the appropriate iodine concentration for the immersion times described above. SEM analysis was to be performed on membrane immersed in iodine for 0, 1, 5, 10, 30, 45, 60, and 83 days. The additional times chosen for sampling were to allow for detection of the earliest possible changes in structural features. Not all concentrations of iodine were examined by SEM at the added time periods (see Results and Appendix B). Multiple membrane pieces (4-6) for each concentration and time were dried in vaccuo at 0.03-0.01 torr for a minimum of twenty-four hours. The specimens were attached to aluminum holding stubs with a conductive adhesive (TV Tube Koat®) and allowed to stand until the adhesive dried (less than 0.5 hours). The stubs containing the membrane specimens were individually coated with gold in an sputter-coater (ISI Model P-S1) at 1.2 kv and 40 ma at 0.1 torr for two minutes (coating thickness approximately 500 Å). Specimens were viewed with an ISI M-7 Scanning Electron Microscope at 15 kv and a 0.7 working distance factor. Magnifications from 1,400 to 14,000 X were examined to determine specimen detail. Micrographs were obtained with a Polaroid CU-5 close-up hand camera (4x5 photography). Polaroid P-N 55 film was used to obtain both positive and negative prints.

SEM analysis was performed on the front and back surfaces of the polyamide membrane. The spacing and other backing materials had been removed as described in the immersion study. As a control, polyamide membrane immersed

in only the buffer solution (0.01M phosphate at pH=6.9) was analyzed by SEM in the same manner as described for the iodine studies.

Disinfectant Residual Analysis - Immersion Studies. For the determination of iodine residuals in the membrane immersion studies the amperometric titration technique, as described in Standard Methods (12), was used with modification of the test solution to pH 7. Standard Methods sets forth the titration to be conducted in a pH 4, acetate buffered solution. However, it was demonstrated that amperometric titrations performed in a pH 7, phosphate buffered medium exhibited no difference in the titration results. A Fischer and Porter amperometric titrator (Model 17T1010) was employed for all amperometric titration analyses.

Selection of Test Organisms. A polyamide membrane was subjected to Potomac River water by USAMERADCOM personnel until the membrane demonstrated a significantly decreased flux rate. The wet membrane was then air-shipped to UTSA where it underwent initial isolation and identification of microorganisms from the fouled membrane.

Membrane preparation was performed under a desk-top hood equipped with a UV light source. Prior to membrane handling, the work area was disinfected with an iodophor wash and subjected to UV irradiation for at least 1 hour. Using aseptic techniques, the membrane was unrolled and 20 cm x 10 cm strips of membrane and supporting materials were cut out. During these manipulations a maximal effort was made to minimize exposure and handling of the membrane surface. The membrane strips were placed in a sterile 250 ml centrifuge bottle and 100 ml of phosphate-buffered saline were added. The centrifuge bottle was placed on a platform shaker and rotated for 1 hour at 200 rpm.

The resulting saline wash was collected aseptically and spread plated in triplicate at various dilutions on several different types of bacteriological

medias. The medias were chosen in an effort to insure the recovery of a diverse number of organism types which might have been present on the membrane. The medias used for organism detection were:

1. Heart infusion agar - a highly enriched medium which is relatively nonselective for organism type.

2. Cetrimide agar - a selective medium for fluorescein-producing Pseudomonas.

3. MacConkey's agar - an enriched medium for a wide variety of gram negative organisms.

4. Eosin methylene blue agar - a nonselective medium for gram negative organisms.

5. Hektoen agar - a moderately selective medium for gram negative organisms.

Following incubation at 37C for varying periods of time (≥ 24 hr.), representative colonies were fished from the plates and cultured on heart infusion slants. Tentative isolate identifications were performed using appropriate biochemical screens. The type of biochemical screen used depended upon the results obtained after subjecting each organism to the oxidase test and the triple sugar iron (TSI) test. Confirmation of bacterial identification was made by use of the Enterotube® and Oxi/Ferm tube® systems. Cultures of the most prevalent organisms were maintained on heart infusion agar slants for future use.

The target organisms chosen for determination of the inactivation efficiency of candidate disinfectants based upon their relative predominance were Pseudomonas fluorescens, Pseudomonas aeruginosa, and Escherichia coli. A further criteria considered in the selection of the Pseudomonas sp. was their relative resistance to disinfection.

Biological Disinfection Systems. For these experiments, a 15 to 18-hour culture of the test organism was grown on a nutrient slant. Organisms were harvested from the slant as a suspension using 10 ml of 0.9% sodium chloride solution (pH=5.5), Travenol. This cell suspension was added to 990 ml of Travenol contained in a sterile two-liter trypsinizing flask and allowed to mix turbulently for 15 minutes to maximize cell dispersion. (See Figure 1 for schematic of disinfection system.) Immediately before the addition of the disinfectant, an aliquot was removed aseptically to determine the initial concentration of organisms in the test system. A titered amount of disinfectant was added into the system under rapid-mix conditions using a syringe. Samples for biological and residual analysis were retrieved at selected contact times over the 30 minute test period. A Cornwall pipette was used for sampling. Samples for bacterial analysis were introduced directly into brain heart infusion (BHI) blanks of known volume. The high organic content of the BHI instantaneously reduced the disinfectant residual.

Analyses for test organisms were carried out by direct plating onto the appropriate selective media allowing enumeration of typical fluorescent or pigmented colonies. Selective medias for the Pseudomonas sp. and Escherichia coli were Cetrimide agar and MacConkey's agar, respectively, overlaid with tryptose phosphate (TPO_4) soft agar into which one ml volumes of appropriate dilutions had been inoculated. Plates were incubated for 24 hours prior to enumeration. Aseptic technique was maintained throughout the test procedure.

Disinfectant Residual Analysis - Biological Studies. The leuco crystal violet (LCV) technique, as described in Standard Methods (12), was adapted for halogen (iodine, iodine bromide, and chlorine) residual determinations for the biological studies. The LCV spectroscopic procedure was adapted for small sample volumes of 5 ml due to the requirements of the biological reactor

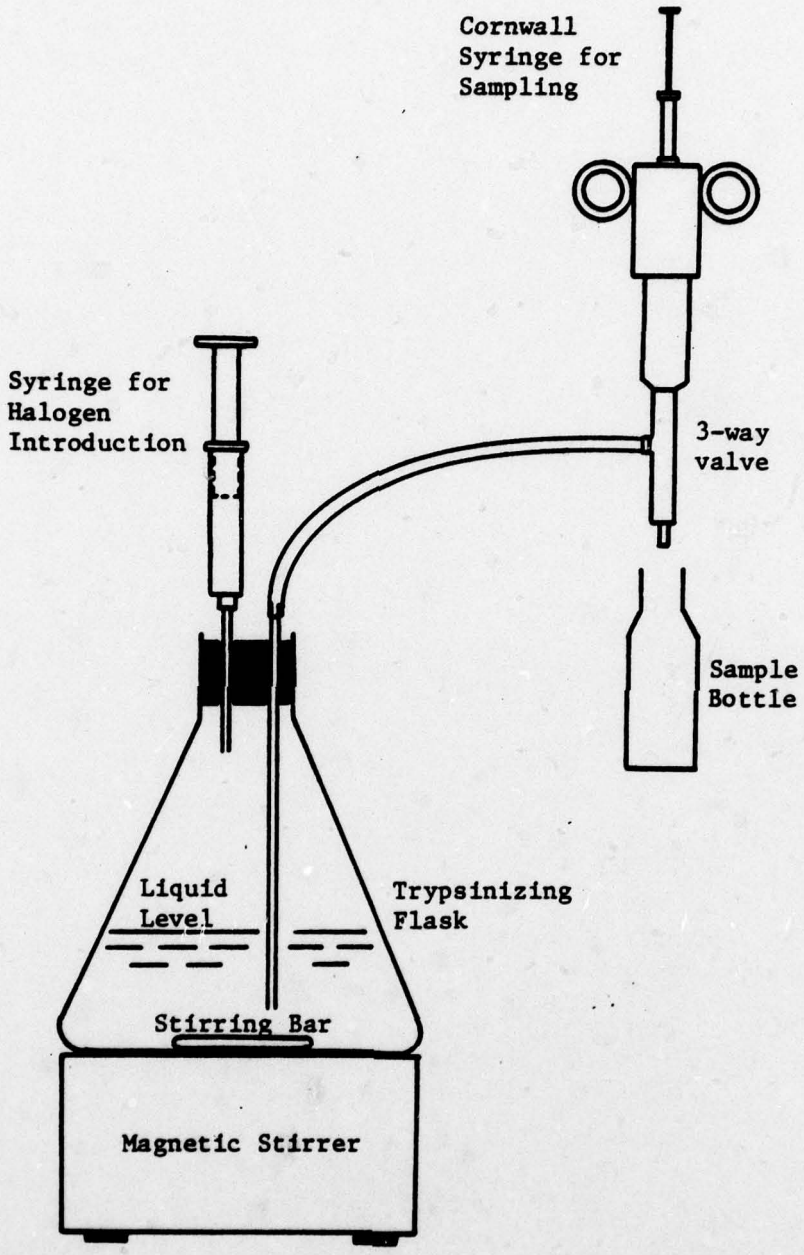


FIGURE 1. Disinfection System

system. A Beckman Century Acta II UV-Vis recording spectrophotometer was used for all LCV measurements at a wavelength of 592 nm. A disadvantage of the LCV technique was that instrument error was minimum at one absorbance unit and increased below that value. Thus, a practical lower limit to detection was approximately 0.0492 mg/l of iodine, 0.186 mg/l of iodine bromide, 0.0441 mg/l of total chlorine and 0.0981 mg/l of free chlorine. A standard curve prepared for the LCV technique using amperometric titration as the referee technique for iodine concentrations between 0.642 mg/l and 6.42 mg/l yielded a regression coefficient of 0.999 indicating extremely good linearity of response in this region. Equation 1 gives the regression line where A_s is the sample absorbance and A_{t_o} is the absorbance at time zero.

$$(I_2) = 6.29\ (A_s - A_{t_o}) + 0.0492 \qquad (1)$$

Iodine concentrations above 6.0 mg/l required dilution with buffered iodine demand-free water to a final concentration of less than 6.0 mg/l. Iodine solutions for the standard curves were prepared according to Standard Methods (12).

The LCV technique as applied to iodine was adapted for use with iodine bromide. Conditions and reagents for the analysis of iodine bromide were the same as for iodine (see Standard Methods (12) for Iodine). Standard solutions of iodine bromide were prepared by dissolving 3 g potassium iodide and the appropriate amount of iodine bromide in demand-free buffered water. Amperometric titration was employed as the referee technique for iodine bromide between 0.5 mg/l and 5 mg/l. The regression line of the calibration curve is given by equation 2 and the regression coefficient was 0.999.

$$(IBr) = 6.03\ (A_s - A_{t_o}) + 0.186 \qquad (2)$$

For both iodine and iodine bromide appropriate volumes of stock solutions were introduced under turbulent mixing conditions into a reactor vessel containing a one liter solution of test organism to produce initial disinfectant concentrations of 10, 5, 3, and 1 mg/l. Stock solutions of 1,000, 500, 300, and 100 mg/l were employed. Five ml samples were removed at 0.25, 1, 2, 5, 15, and 30 minutes after addition of iodine or iodine bromide and analyzed for residual iodine or iodine bromide by comparison to the calibration curves (Equation 1 or 2).

Total chlorine residual was measured by the LCV technique as described in Standard Methods (12). A calibration curve constructed with amperometric titration as the referee technique yielded the regression line given by equation 3 with a regression coefficient of 0.999.

$$(Cl_2)_{Total} = 1.70\ (A_s - A_{t_o}) + 0.0441 \qquad (3)$$

Chlorine concentrations above 2.0 mg/l required dilution with buffered chlorine demand-free water to a final concentration of less than 2.0 mg/l. Appropriate volumes of 1000, 500, 300, and 100 mg/l stock solutions of chlorine were introduced into a reactor vessel as described for iodine and iodine bromide. Five ml samples were withdrawn for the 1 and 3 mg/l doses; however, two ml samples were removed for the 5 and 10 mg/l doses. The times of sample withdrawal were the same as for iodine and iodine bromide. Total residual chlorine was then measured by comparison to the standard calibration curves (Equation 3).

Free chlorine was also measured by the LCV technique described in Standard Methods (12). A standard curve constructed with amperometric titration as the referee technique yielded the regression line given by equation 4 with a regression coefficient of 0.999.

$$(Cl_2)_{Free} = 5.60\ (A_s - A_{t_o}) + 0.0981 \qquad (4)$$

The biological reactor was dosed as described for iodine, iodine bromide, and total chlorine. Five ml samples were withdrawn at 0.25, 0.75, 1.75, 3.75, 5.75, and 7.75 minutes. Free chlorine was determined by comparison to the standard calibration curve (Equation 4).

RESULTS

Preliminary Immersion Studies. Initial protocol development led quickly to the restructuring of test procedures. Demand-free Nalgene vessels were planned for immersion tests; however, approximately 80% loss of the initial iodine (or iodine bromide) concentration in three days was observed. Furthermore, rates of iodine loss were dependent upon the amount of head space (gas area) above the liquid level. Increasing the head space by withdrawing constant aliquots (200 ml) at equal time intervals for amperometric determination of iodine concentration showed a loss rate of 0.53 mg/day. On the other hand, demand-free glass chromatography tanks with a flat cover and a small head space (0.25 in.) showed only a 1.7% decrease in total iodine concentration over seven-day trial periods.

Control experiments to determine if stratification of the iodine solutions occurred, especially at high iodine concentrations, were negative. Therefore, no mechanical mixing was employed in these systems. Control experiments were performed to demonstrate the effect of buffer (pH=6.9) on the polyamide and polycellulose acetate membranes. Seven-day trial experiments at various phosphate buffer concentrations showed neither a pH drop nor any qualitative visual effect on the membrane. These and other experiments allowed validation of the amperometric titration procedure at a pH of 7 rather than at a pH of 4 as per Standard Methods (12).

Iodine or iodine bromide uptake or absorption was demonstrated for the polyamide and polycellulose acetate membranes as well as the individual components that comprise the complete membrane such as the backing and spacer. Table 1 shows the absorption of iodine by the polyamide membrane components immersed in a 6 mg/l iodine solution. Similar results were observed with

iodine bromide and the polycellulose acetate membrane. Furthermore, it was found that buffering of the solution at pH of 6.9-7.0 (phosphate, 0.002-0.1M) increases the iodine demand of the polyamide and polycellulose acetate membranes over unbuffered media.

Table 1. 24-Hour Immersion Test of Polyamide Membrane Module Components (Glass Reaction Vessels)*

Component	Change in Iodine Concentration (mg/1)
Porous Backing	-0.427
Mesh Spacer	-0.045
Membrane	-1.289

*No buffer present; pH of solutions was ca. 7.

The final protocol for the membrane immersion studies resulted from initial week-long trials of polyamide and polycellulose acetate membranes immersed in 6.1 mg/1 iodine and 7.5 mg/1 iodine bromide. Figure 2 shows the disinfectant effect on polyamide and polycellulose acetate membranes. The polycellulose acetate membrane had an accelerated disinfectant uptake when compared to the polyamide membrane. Based on this and other considerations the polycellulose acetate membrane was dropped from further study. Comparison of the iodine and iodine bromide effects on the polyamide membrane immersion show similar results (Figure 2). Iodine bromide was absorbed at a three-fold slower rate than was observed for iodine. Discoloration of the membrane occurred with both disinfectants during the test time; however, the iodine bromide treated membrane was considerably lighter in color. Since iodine and iodine bromide behaved similarly in the immersion study and since it was

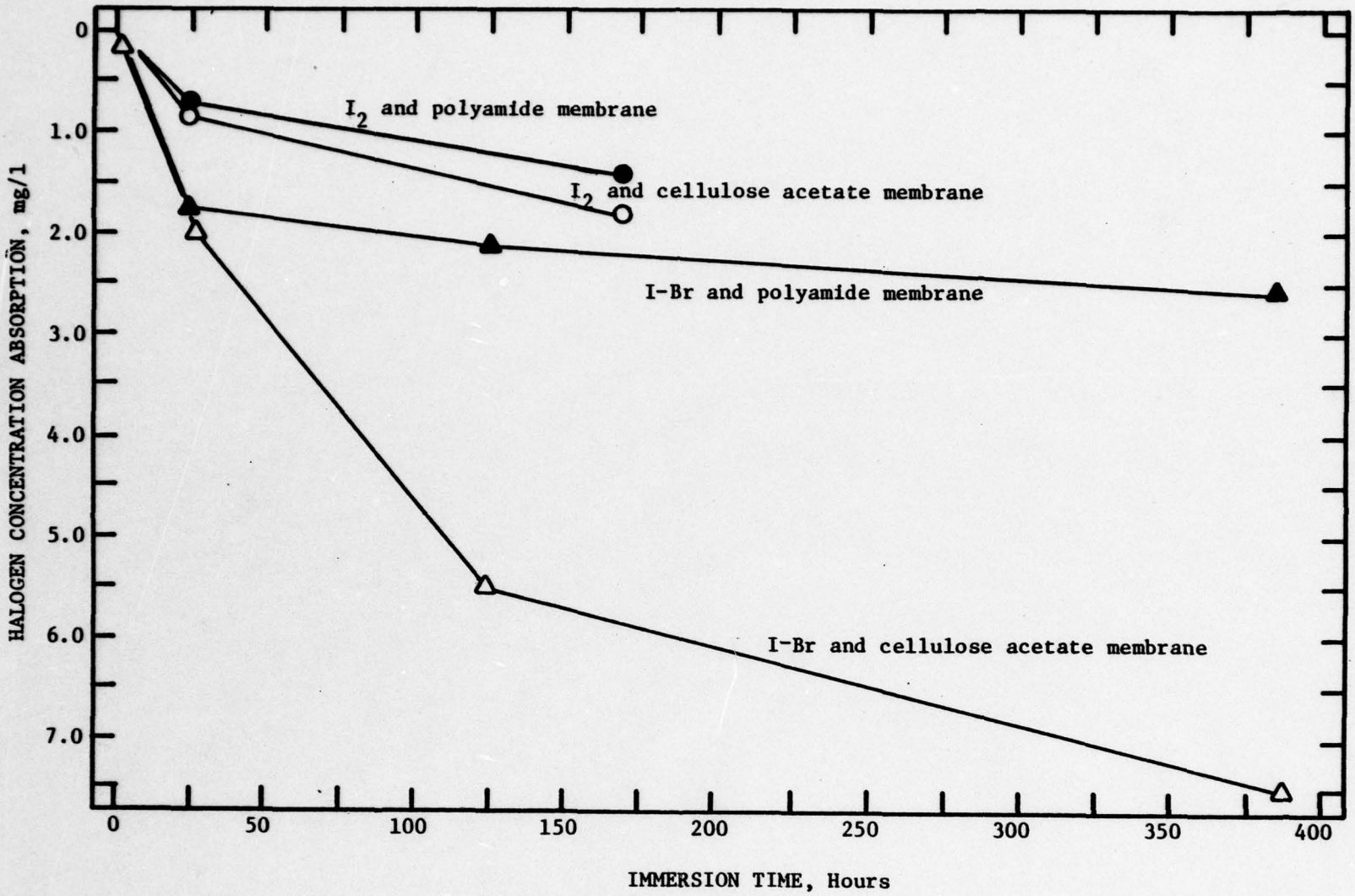


FIGURE 2. Absorption of iodine and iodine bromide by cellulose acetate and polyamide membranes at pH = 7.0.

necessary to perform a labor intensive experiment to keep a relatively constant concentration of disinfectant, iodine only was examined in the complete 2000-hour immersion study. The iodine bromide would be expected to behave qualitatively the same as iodine at 1 and 5 mg/l, but at a slightly slower rate. At high disinfectant concentrations no rate differences were apparent.

Dibromamine was a disinfectant of choice in the early planning stages. Control experiments demonstrated the rapid decomposition of the dibromamine species under the experimental conditions utilized in the immersion study. A protocol for amperometric detection of dibromamine was developed. At an initial concentration of dibromamine of 3.84 mg/l the half-life of the dibromamine was 37 minutes and at a concentration of 1.35 mg/l the half-life was 55 minutes. Since one half of the initial concentration was gone in 37 minutes, at a relatively low concentration, it was not feasible to perform any membrane immersion studies with dibromamine as the disinfectant in the static tank test system. A continuous-flow system would have been necessary to maintain an acceptable dibromamine concentration excursion.

Control experiments have demonstrated that an iodine concentration excursion of less than ±10% was necessary to evaluate the effect of iodine upon the membrane. This required head space minimization to reduce volatilization, all glass vessels or tanks, light exclusion, demand-free buffered solutions, pH control of solution, and frequent solution exchange.

Polyamide Membrane Immersion Studies. The 2000-hour immersion studies with iodine concentrations of 1, 5, and 50 mg/l all had frequent solution changes and the observed excursions did not exceed ±5%. The foil covered, glass topped, chromatography tanks presented no problems except for the large amount of solutions necessary to perform the experiment. Control experiments to determine the maximum intrinsic iodine loss between solution changes (head

space volatilization) for the experimental system showed small changes. With a 1 mg/l concentration of iodine the maximal time between changes was 24 hours during the 2000-hour run. The intrinsic iodine loss of the solution (no membrane) was 0.0026 mg/l/hour. The 5 mg/l iodine concentration allowed a maximal time between changes toward the end of the 2000-hour period of 45 hours. The intrinsic iodine loss was maximally 0.0086 mg/l/hr. Similarly, the 50 mg/l iodine concentration allowed an even longer time between changes (168 hours). The intrinsic iodine loss was maximally 0.0470 mg/l/hr. Tables A-1, A-2 and A-3, Appendix A, present the results for the 1 mg/l, 5 mg/l and 50 mg/l respectively, iodine concentration 2000-hour immersion study. These data were corrected for the intrinsic iodine loss.

Visually the membrane pieces became reddish-brown with exposure to the iodine solutions. The membrane dosed at 1 mg/l was the slowest to change and was light reddish-brown at the end of the study. The membrane dosed at 5 mg/l turned dark quickly and the membrane dosed at 50 mg/l was deep reddish-brown after several hours. The color changes are a function of iodine uptake as reflected in the tabulated data. The membrane immersed at 1 mg/l iodine concentration absorbed a total of 8 mg/l of iodine, the membrane immersed at 5 mg/l absorbed a total of 30 mg/l and the membrane immersed at 50 mg/l absorbed a total of 59 mg/l. No visual deterioration of the membrane surface was detectable at any of the test concentrations. All membrane immersion studies show an apparent reduction of iodine uptake at prolonged immersion times (See Appendix A). However, a relatively constant rate of iodine absorption continued throughout the 2000-hour study.

SEM Analysis. The cut membrane strips prepared for the SEM analysis showed no changes upon wet storage in glass vials that were protected from light. Drying of the membrane strips was necessary for gold coating and

subsequent SEM analysis. Control experiments showed no differences between vacuum-dried, air-dried, virgin, and buffer-treated membrane strips under SEM analysis.

Scanning electron micrographs of the 0, 1, 5, and 50 mg/l iodine treated membrane pieces at magnifications of 1,400-14,000 X for selected times are contained in Appendix B.

Changes in the structural appearance of the polyamide membrane after immersion in iodine solutions or the buffer alone were detectable by scanning electron microscopy. In general, the changes are sequentially related to the time of immersion and to the concentration of iodine present in the test system. Furthermore, the observed changes occurred on both the front and back sides of the polyamide membrane strip (Appendix B).

Figures 3, 4, 5 and 6 show the effect of phosphate buffer (0.01M, pH=6.9), and iodine concentration of 1, 5, and 50 mg/l, respectively, on the membrane surface at times of 0, 5, 45, and 83 days. All time zero photographs (Figures 3A, 4A, 5A, and 6A) show a surface that is essentially identical when examined under the same conditions (magnification 14,000 x). The membrane (front and back) was characterized by small light nodules that are distinct and well formed. The modification of these nodules was interpreted as a structural change in the membrane. The approximate size of these nodules for the membrane front is presented in Table 2. No other structural features, such as holes or cracks, were observed at time zero.

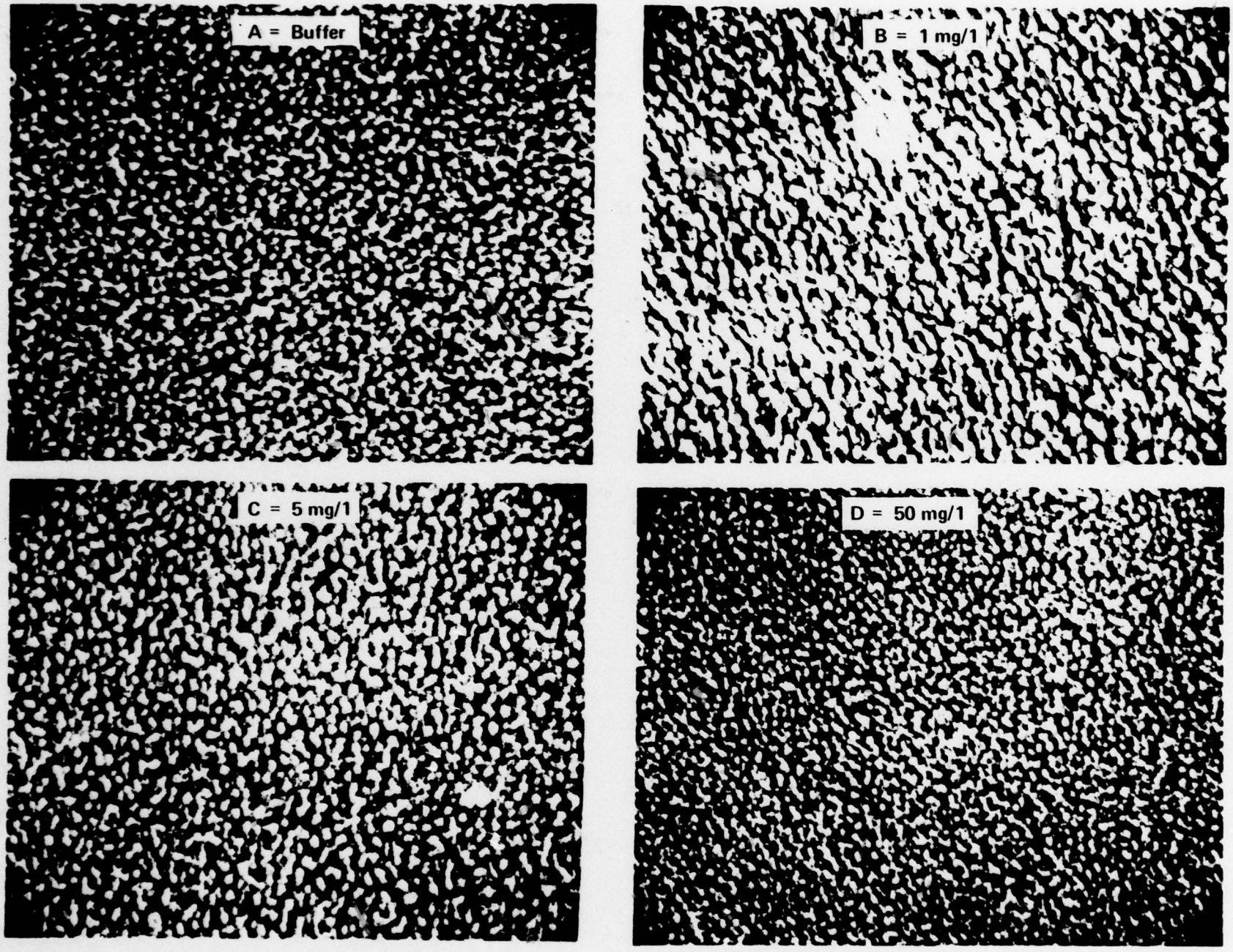


FIGURE 3. Scanning Electron Micrographs of Polyamide Membrane at Time = 0 Days (14,000x)

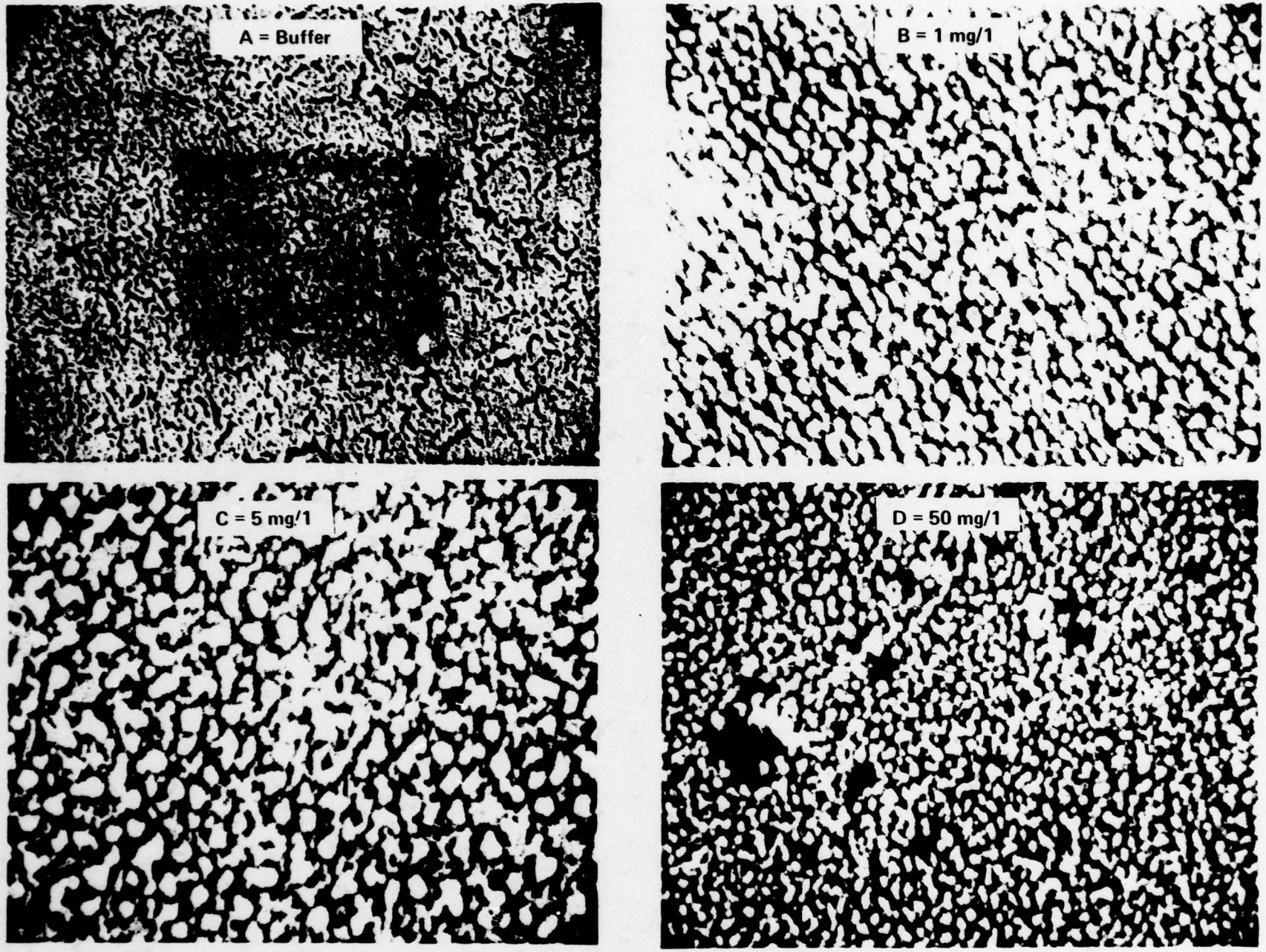


FIGURE 4. Scanning Electron Micrographs of Polyamide Membrane at Time = 5 Days (14,000x)

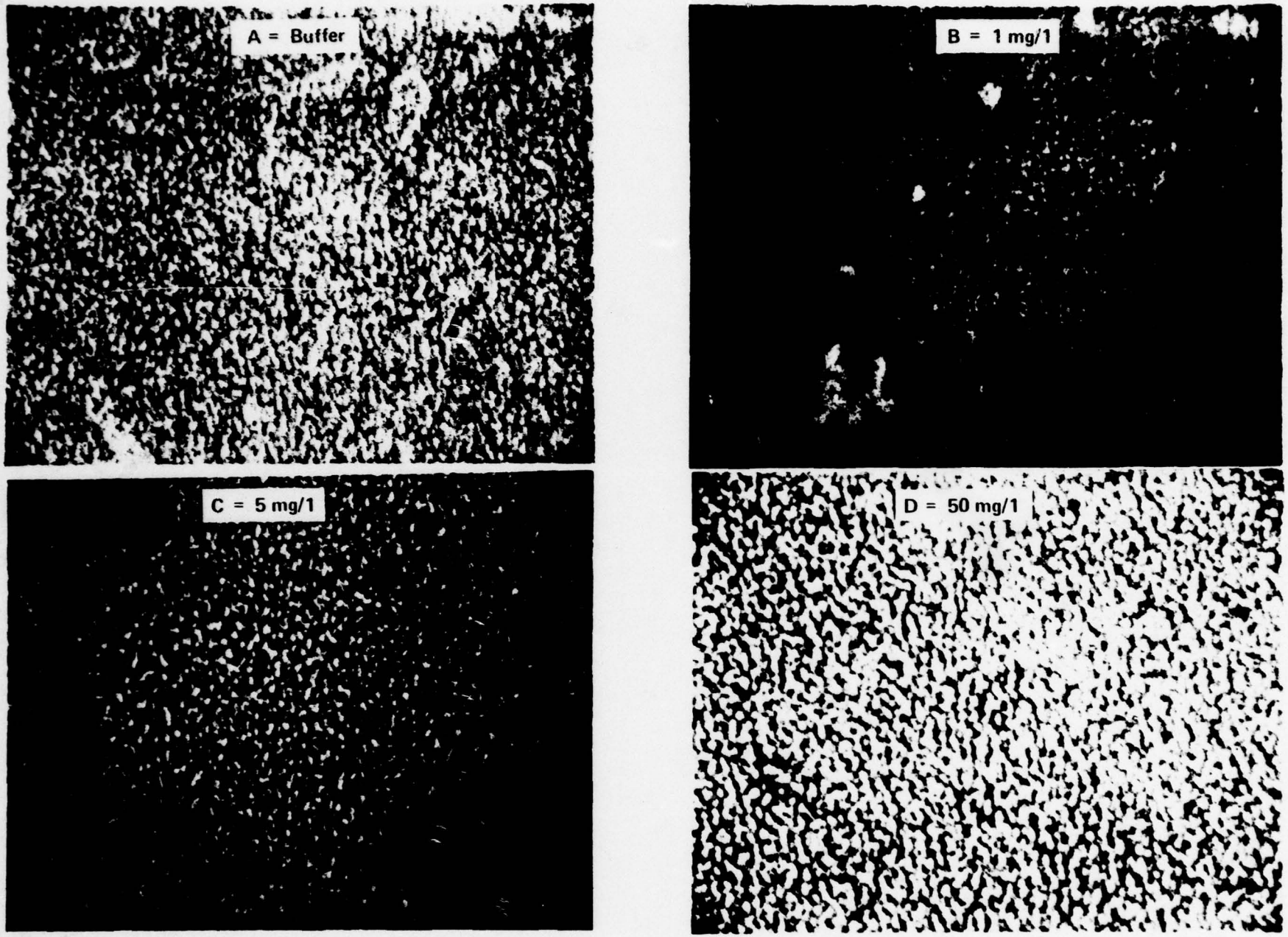


FIGURE 5. Scanning Electron Micrographs of Polyamide Membrane at Time = 45 Days (14,000x)

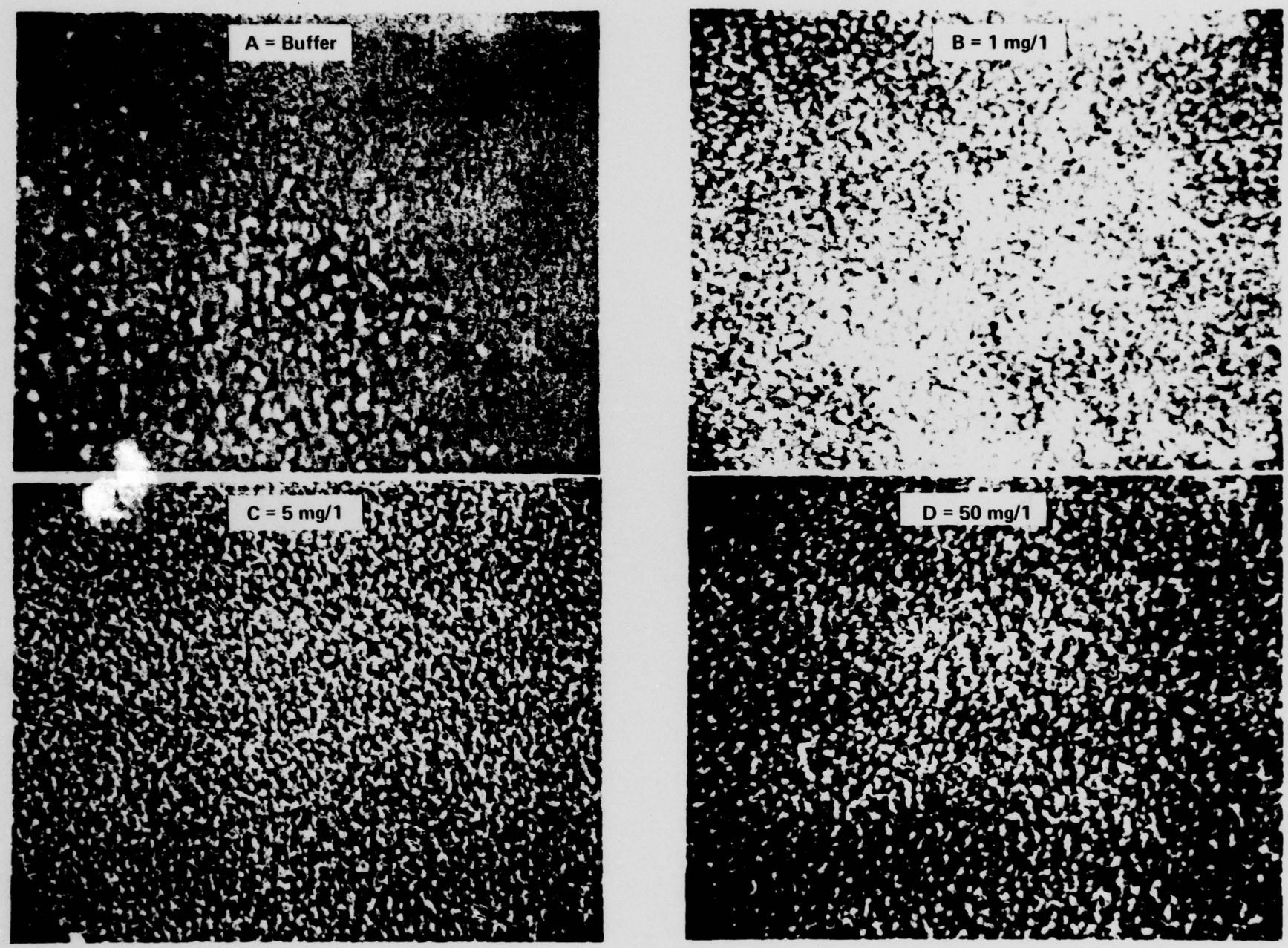


FIGURE 6. Scanning Electron Micrographs of Polyamide Membrane at Time = 83 Days (14,000x)

Table 2. Size of Nodules and Holes in Polyamide Membrane Immersed in Iodine

Type	Time (days)	Approximate Size in Angstroms (averaged values) 0	1 mg/l	5 mg/l	50 mg/l
Nodules	0	1430	1430	1430	1070
	5	*	1790	2860	1250
	45	710	710	710	820
	83	930	710	710	780
Holes	0	†	†	†	†
	5	†	†	†	3570
	45	†	1430	2500	3570
	83	†	1430	2500	3570

*See text; † No observed holes.

Examination of the micrographs for the membrane immersed only in buffer (Figure 3) exhibited some small changes. Figure 3B is anomalous and cannot be explained at this time. Micrographs at 1 and 10 days (Appendix B) are essentially the same as the 45 day picture (Figure 3C). The nodules shrink with prolonged exposure to the buffer, and the 83 day exposure (Figure 3D) was essentially the same as the iodine immersion experiments at all iodine concentrations. No other structural features, except the nodules, were apparent from the micrographs.

The 1 mg/l and 5 mg/l iodine concentration immersion runs behaved in a similar manner (Figures 4 and 5). The initial nodules increased in size during the first five days. The 5 mg/l iodine concentration experiment showed nodules that increased in size about 1.5 times, when compared to the 1 mg/l iodine concentration experiment. With prolonged immersion the nodules then decreased in size with the concomitant development of holes or larger areas

between the small nodules (Table 2). The holes observed for the 1 mg/l iodine concentration were approximately half as small as the holes observed for the 5 mg/l iodine concentration. Continued immersion until the 83 day termination did not affect the size or number of the holes observed.

The 50 mg/l iodine concentration affected the membrane most severely (Figure 6). The changes observed were qualitatively the same as seen for the 1 mg/l and 5 mg/l concentration experiments. However, the changes in membrane structure were accelerated in time. The same process of nodule swelling and shrinkage with concomitant appearance of holes occurred by the fifth day compared to the 45th day for the lower iodine concentrations. Although the holes that appeared were generally of similar size (Figures 6C and D), a very great structural deviation could be observed in the 5 day 50 mg/l iodine immersion study (Figure 6B). A hole with dimensions of 11,430 Å by 12,140 Å was apparent. This was the only sample with such a large hole, but other membrane strips not examined may show similar structural changes. Holes of consistent size and appearance were present in all other membrane samples contacted with the 50 mg/l iodine concentration (Table 2).

On all samples small dust particles were observed. Similar changes in the surface features of the backing material occurred and correlation with the front of the membrane was observed.

Disinfection Studies. The bacterial efficiencies of iodine, iodine bromide, and chlorine for the test organisms Pseudomonas aeruginosa, Pseudomonas fluorescens, and Escherichia coli are shown in Figures 7 through 15. All curves were fitted using bivariate regression analysis and an exponential model such that:

$$\log_{10} Y = Y_0 + bt \qquad (5)$$

where; Y is the surviving bacterial fraction, Y_0 is the intercept (ideally 1.00 for the first disinfection phase), b is the slope (regression coefficient of Y on t), and t is the contact time. The halogen doses shown in Figures 7 through 15 were 1, 3, 5, and 10 mg/1. The temperature and pH of the test solutions were 25±1C and 6.9±0.2, respectively.

Figure 7A shows the bacterial efficiency of iodine for P. aeruginosa. Approximately six logs of inactivation were achieved by two minutes for the 10 mg/1 iodine dose. Whereas, for the lowest dose of 1 mg/1, two logs of inactivation were achieved within two minutes with no significant increase in inactivation occurring during the remainder of the 30 minute contact period. Results intermediate to those obtained for iodine doses of 1 and 10 mg/1 were obtained for iodine doses of 3 and 5 mg/1. All curves are typically biphasic and are characterized by an initial phase of approximately one minute during which inactivation proceeds very rapidly. During the second phase bacterial inactivation ceases or proceeds at a reduced rate.

Iodine residual curves shown in Figure 7B are also biphasic with a high initial rate of iodine demand occurring over a time period corresponding to the initial high rate of P. aeruginosa inactivation. For those iodine doses (3, 5, and 10 mg/1), in which a significant residual remained in the test system following the initial iodine demand, a significant but greatly decreased rate of iodine demand was present during the second stage. The amount of initial iodine demand and total iodine demand during 30 minutes of contact time varied directly as a function of the initial iodine dose.

The results obtained reacting iodine bromide as the disinfectant with P. aeruginosa, as shown in Figure 8A, were similar to the results obtained using iodine as the disinfectant. However, for a 10 mg/1 iodine bromide dose (as iodine) approximately 13.5 minutes of contact time were required to acheive

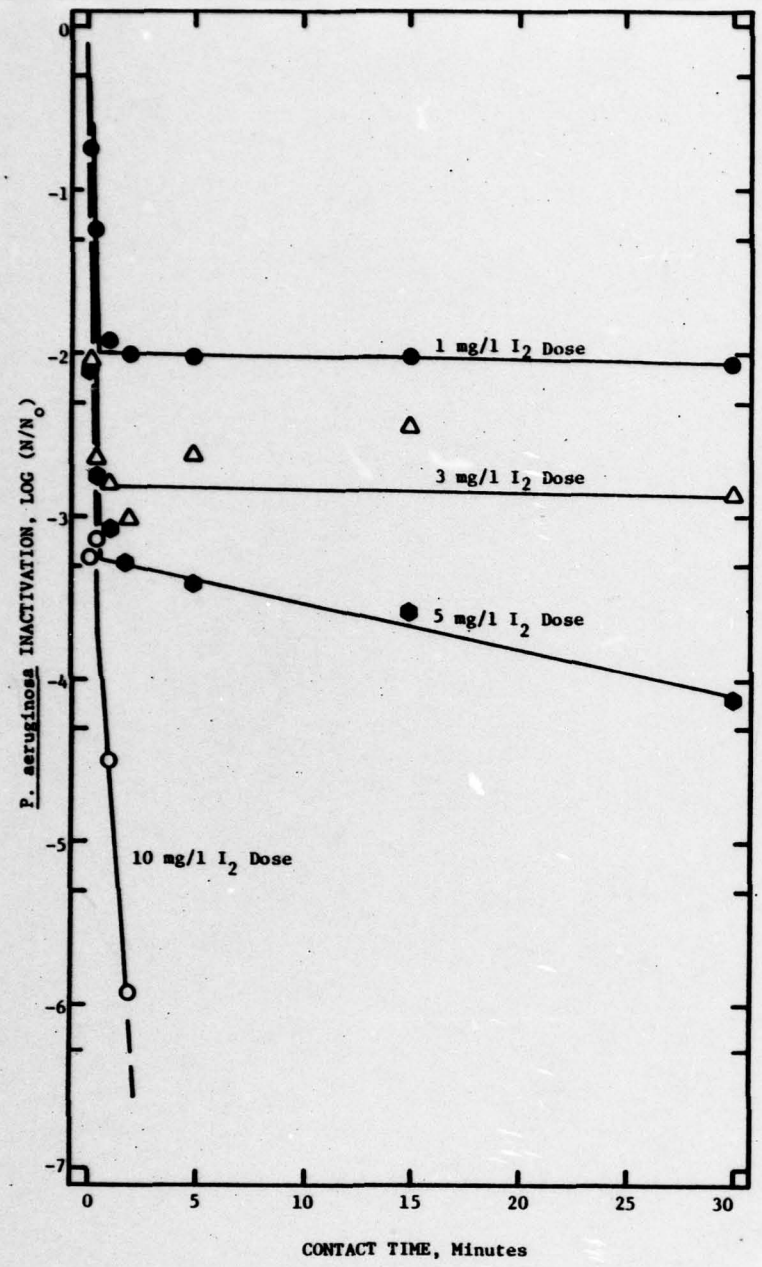


FIGURE 7A. P. aeruginosa inactivation as a function of iodine dose and contact time at 25 ±1C and pH 6.9 ± 0.2.

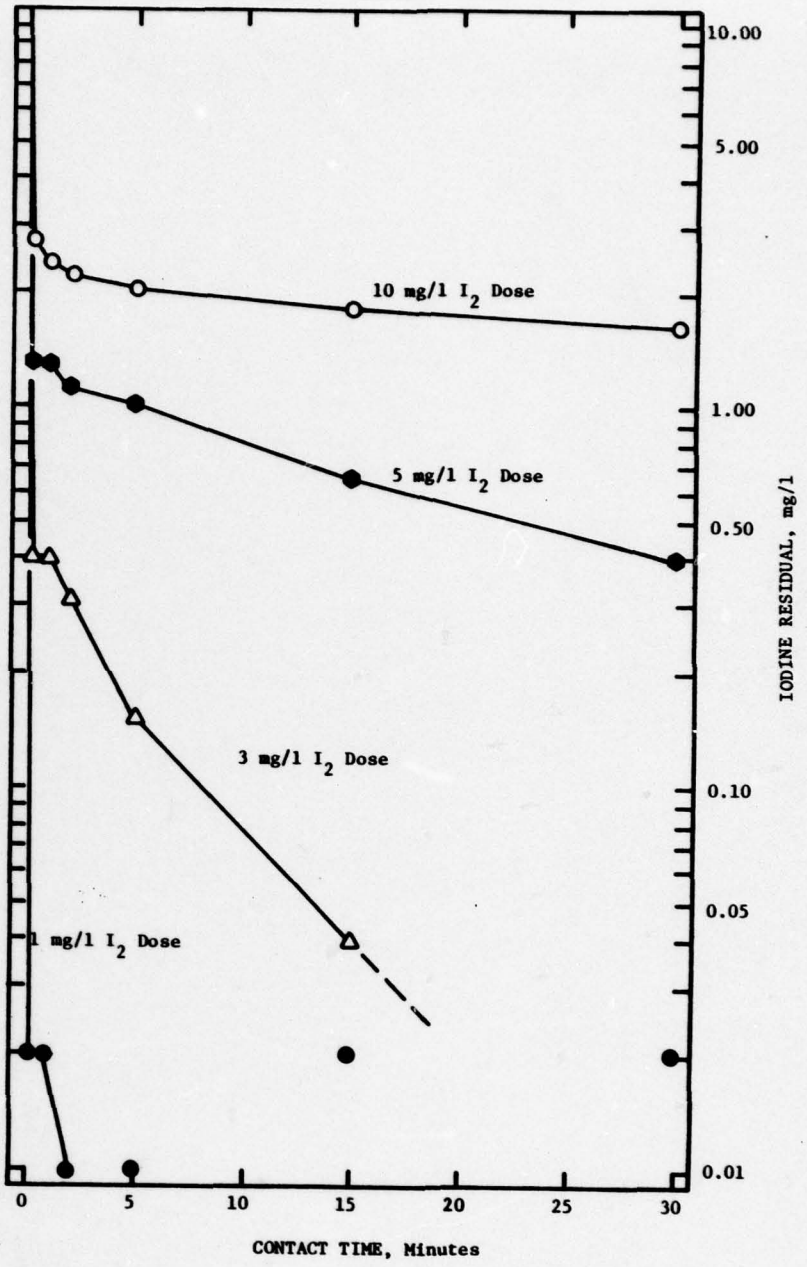


FIGURE 7B. Iodine residual as I_2 as a function of iodine dose and contact time for P. aeruginosa disinfection studies.

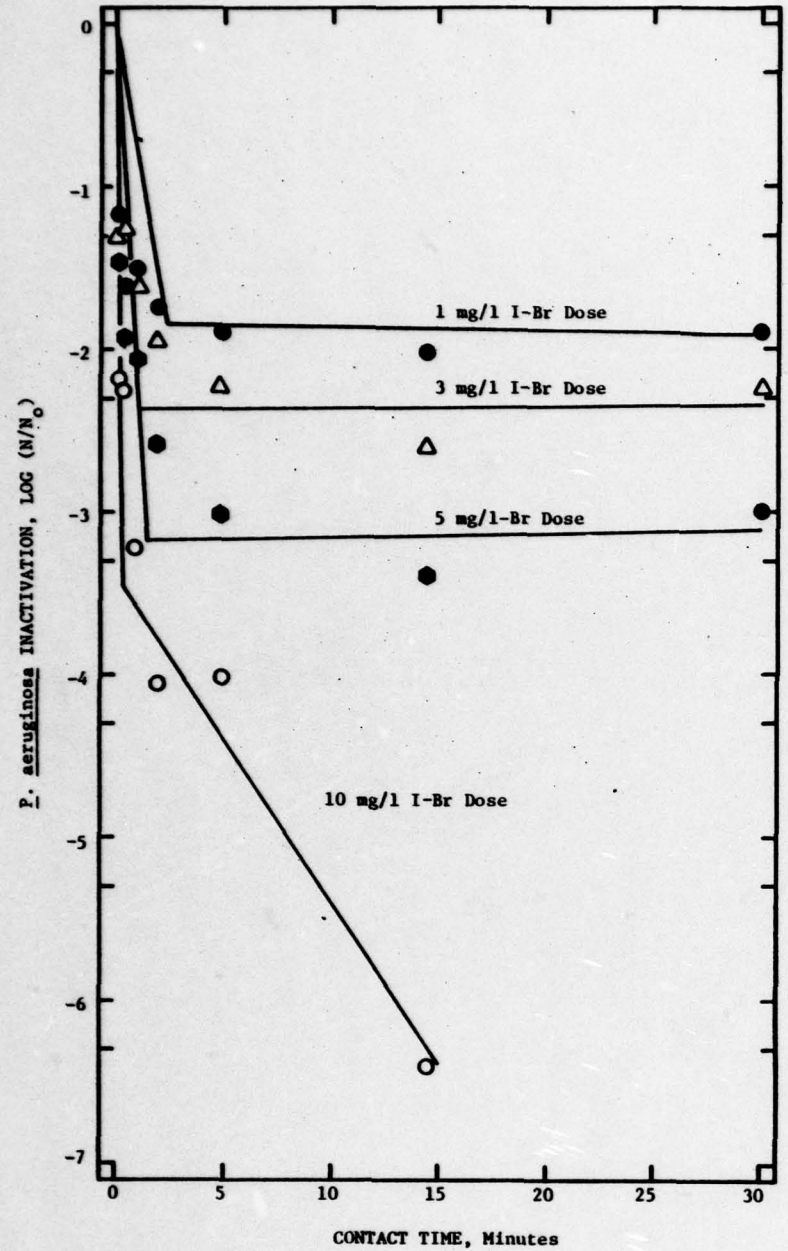


FIGURE 8A. P. aeruginosa inactivation as a function of iodine bromide dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

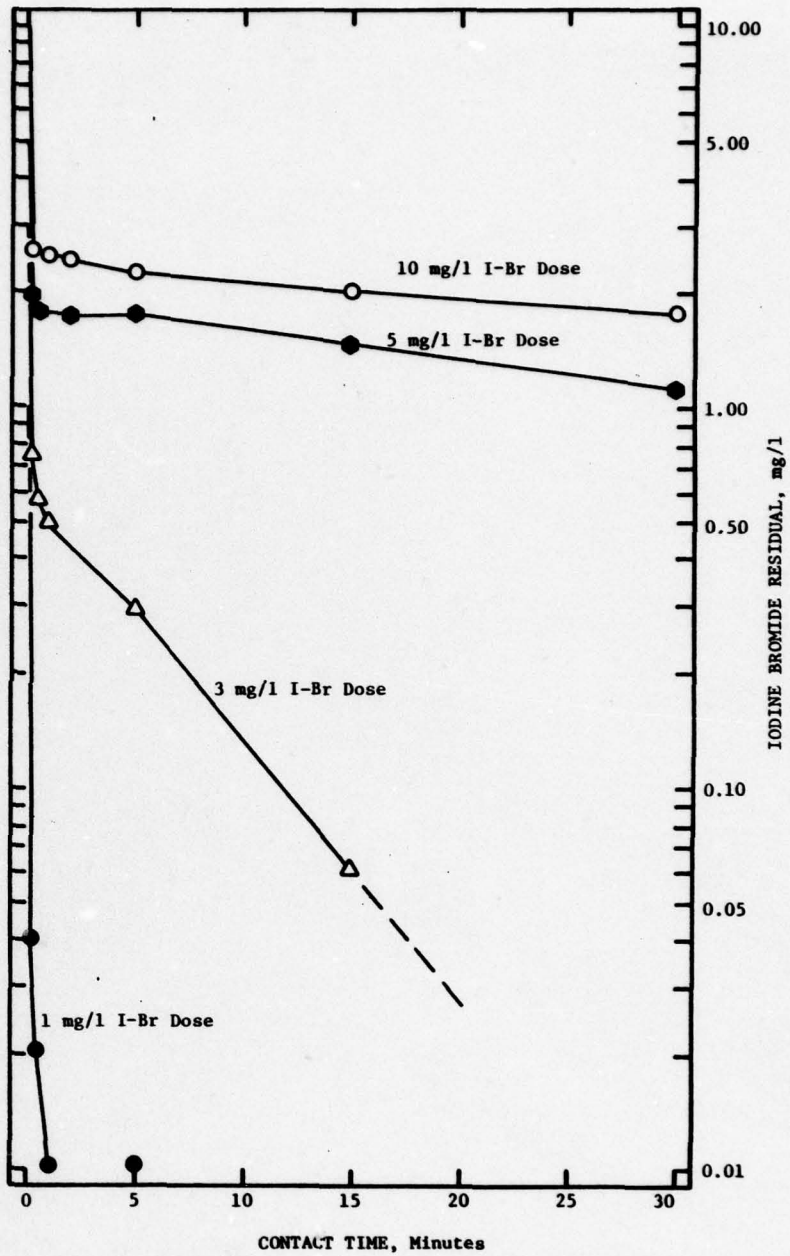


FIGURE 8B. Iodine bromide residual as I_2 as a function of iodine bromide dose and contact time for P. aeruginosa disinfection studies.

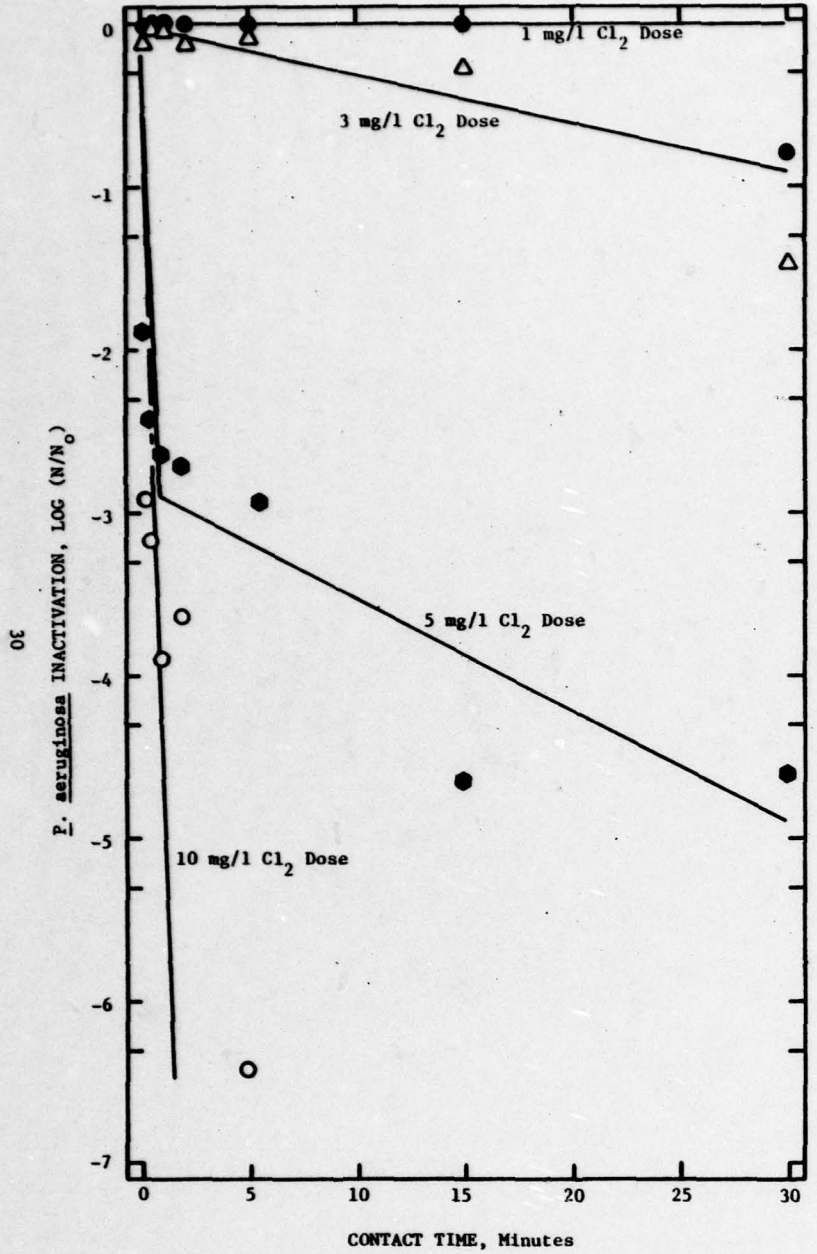


FIGURE 9A. P. aeruginosa inactivation as a function of total chlorine dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

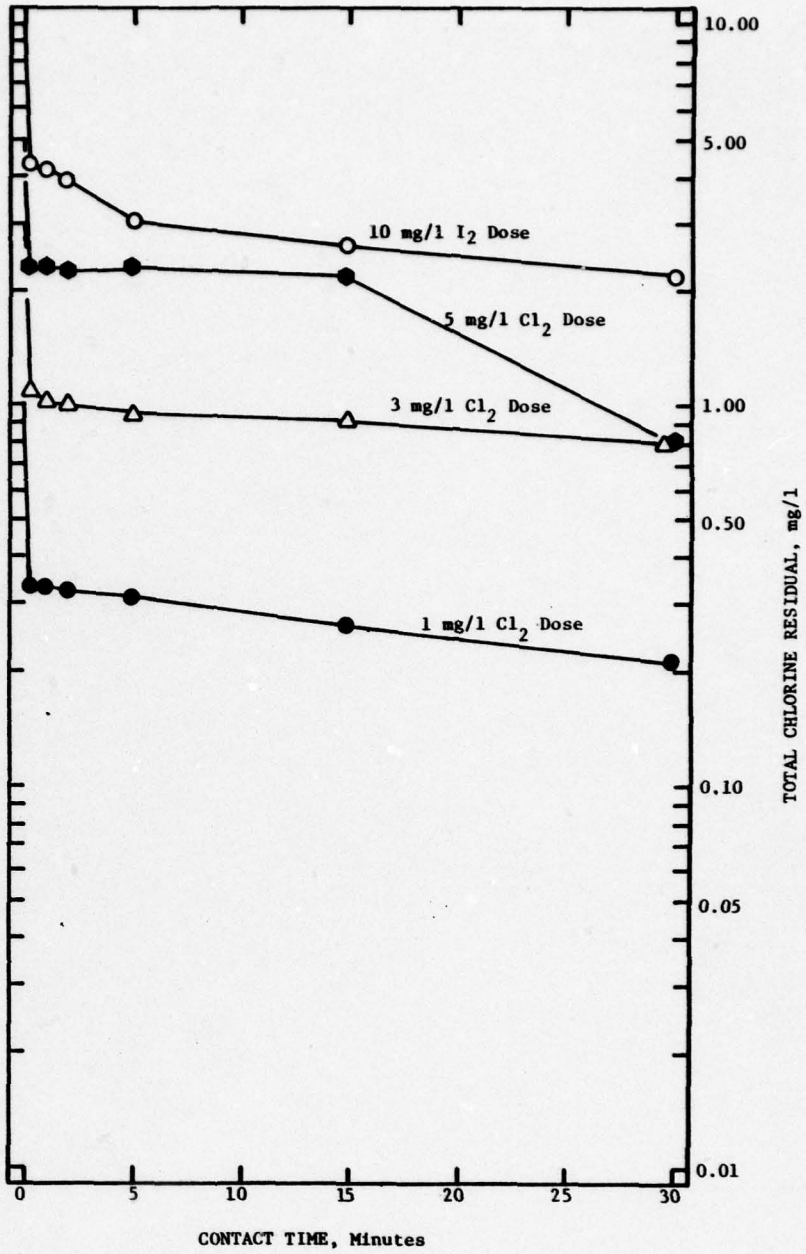


FIGURE 9B. Total chlorine residual as Cl_2 as a function of total chlorine dose and contact time for P. aeruginosa disinfection studies.

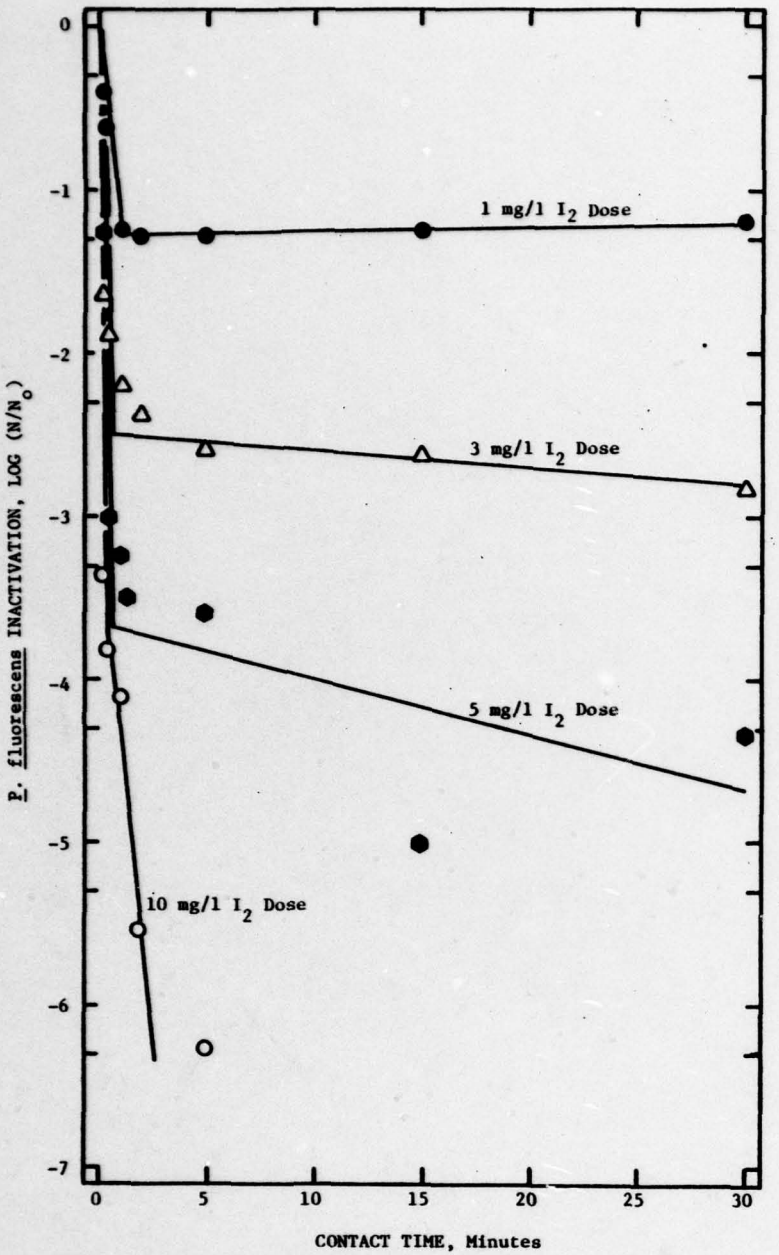


FIGURE 10A. P. fluorescens inactivation as a function of iodine dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

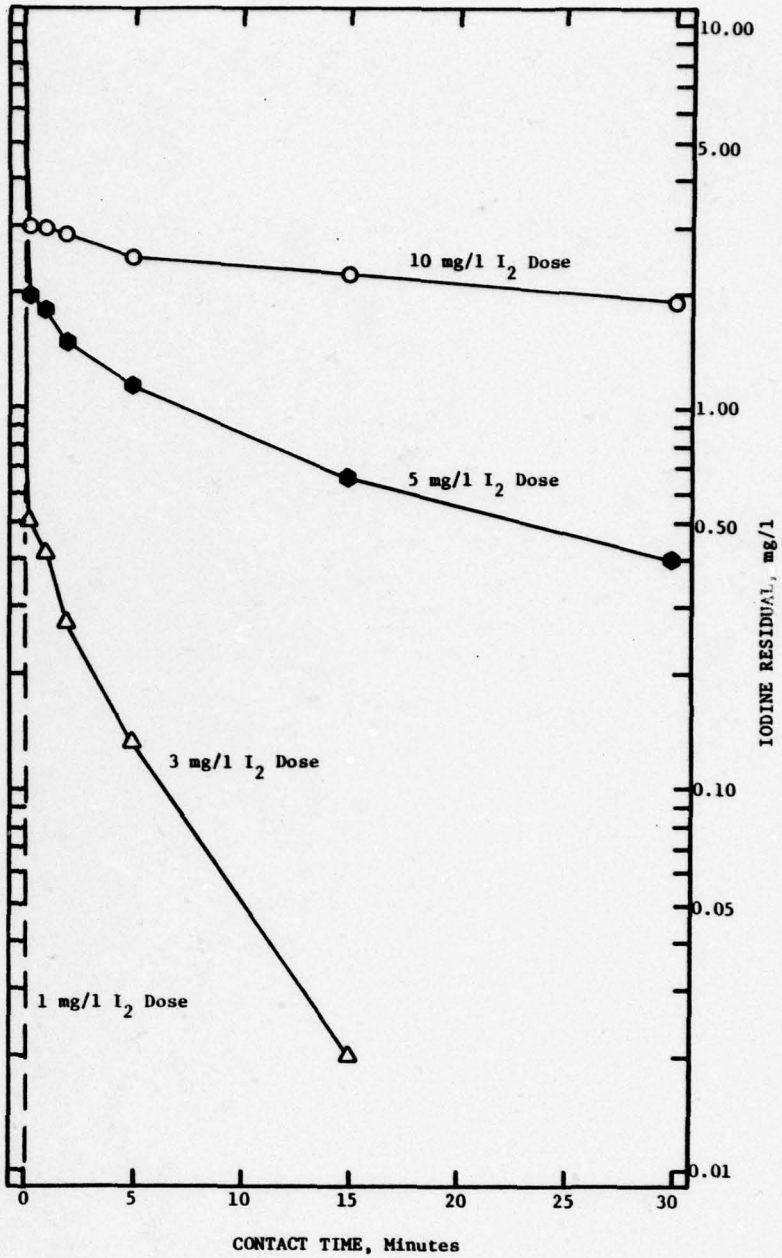


FIGURE 10B. Iodine residual as I_2 as a function of iodine dose and contact time for P. fluorescens disinfection studies.

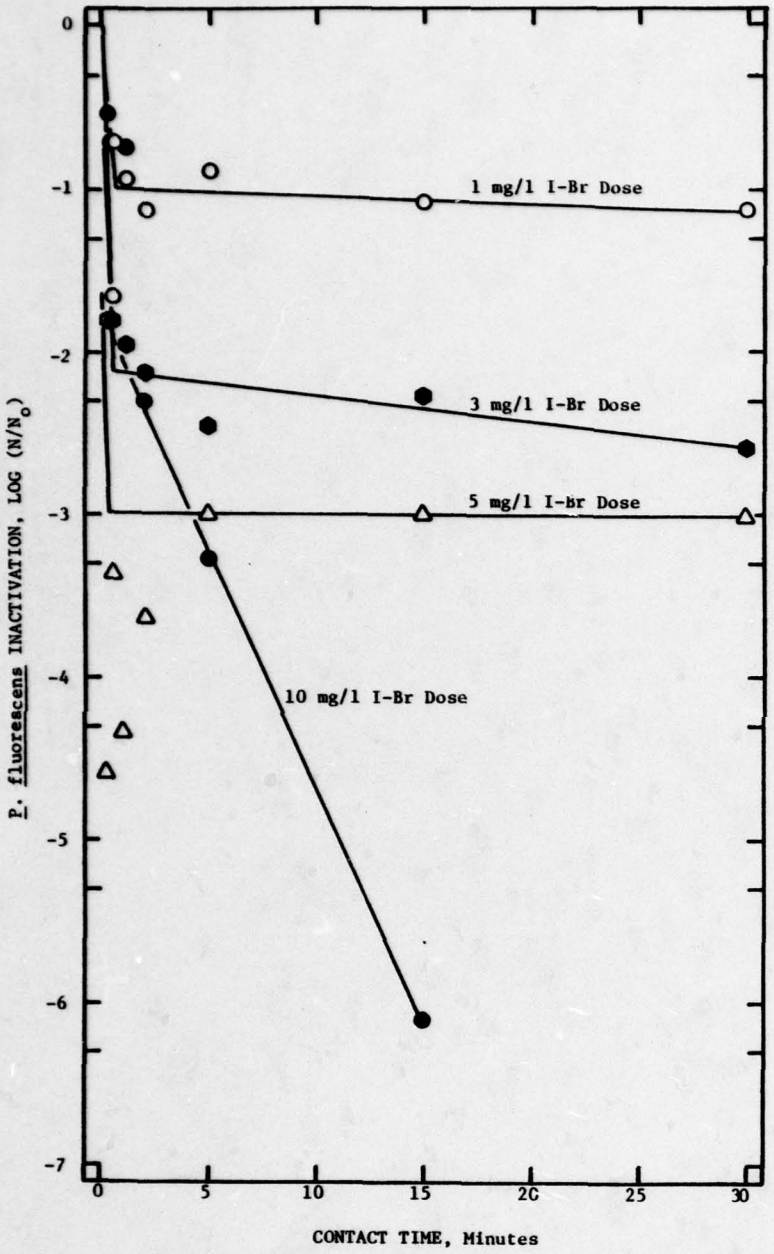


FIGURE 11A. P. fluorescens inactivation as a function of iodine bromide dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

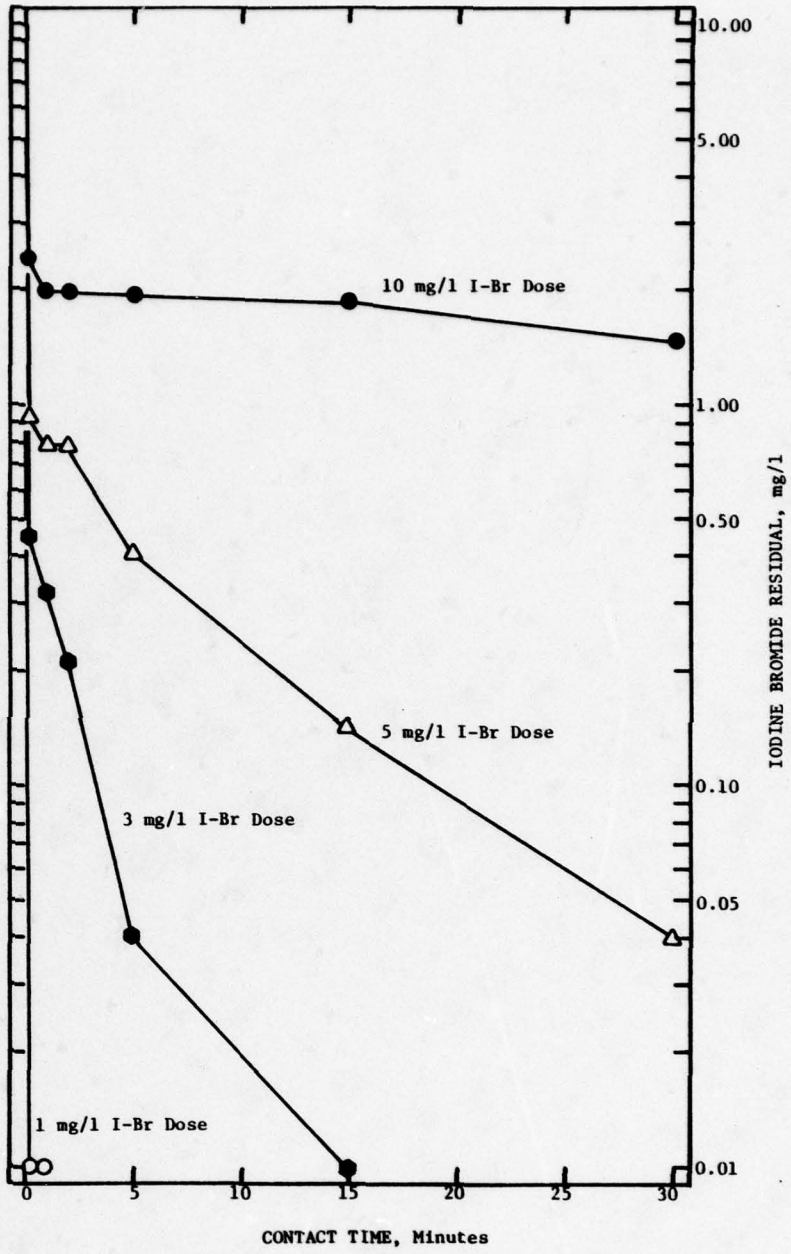


FIGURE 11B. Iodine bromide residual as I_2 as a function of iodine bromide dose and contact time for P. fluorescens disinfection studies.

six logs of inactivation. Results obtained for iodine bromide residual were also similar to those previously described for iodine residual (Figure 8B).

The results obtained by inactivating P. aeruginosa using chlorine, as shown in Figure 9A, were similar to those described above for iodine and iodine bromide when halogen doses of 5 and 10 mg/l were used. However, the data for chlorine doses of 1 and 3 mg/l show remarkably less inactivation than that achieved with corresponding iodine and iodine bromide doses. Approximately one log of P. aeruginosa inactivation was achieved for a chlorine dose of 3 mg/l and 30 minutes contact time. No P. aeruginosa inactivation was demonstrated for a chlorine dose of 1.0 mg/l and 30 minutes contact time. The chlorine residual curves shown on Figure 9B are typically biphasic and demonstrate an initial, rapid chlorine demand with a persisting, but slower rate of chlorine demand becoming established after approximately one minute of contact time and persisting throughout the remaining 30 minutes of contact time. Free chlorine was detectable for less than fifteen seconds with an initial dose of 10 mg/l.

The inactivation and halogen residual data for P. fluorescens using iodine and iodine bromide individually as disinfectants, as shown on Figures 10 and 11 is quite similar to the P. aeruginosa inactivation data using the same disinfectants and presented on Figures 7 and 8, except P. fluorescens appears to be slightly more resistant than P. aeruginosa to inactivation by iodine or iodine bromide.

The data in Figure 12A showing the inactivation of P. fluorescens using chlorine as the disinfectant demonstrates approximately 0.85 log of inactivation for a chlorine dose of 1 mg/l and 30 minutes contact time. Extension of the curves for chlorine doses of 3 and 5 mg/l show greater than seven logs inactivation of P. fluorescens after 30 minutes of contact time. And,

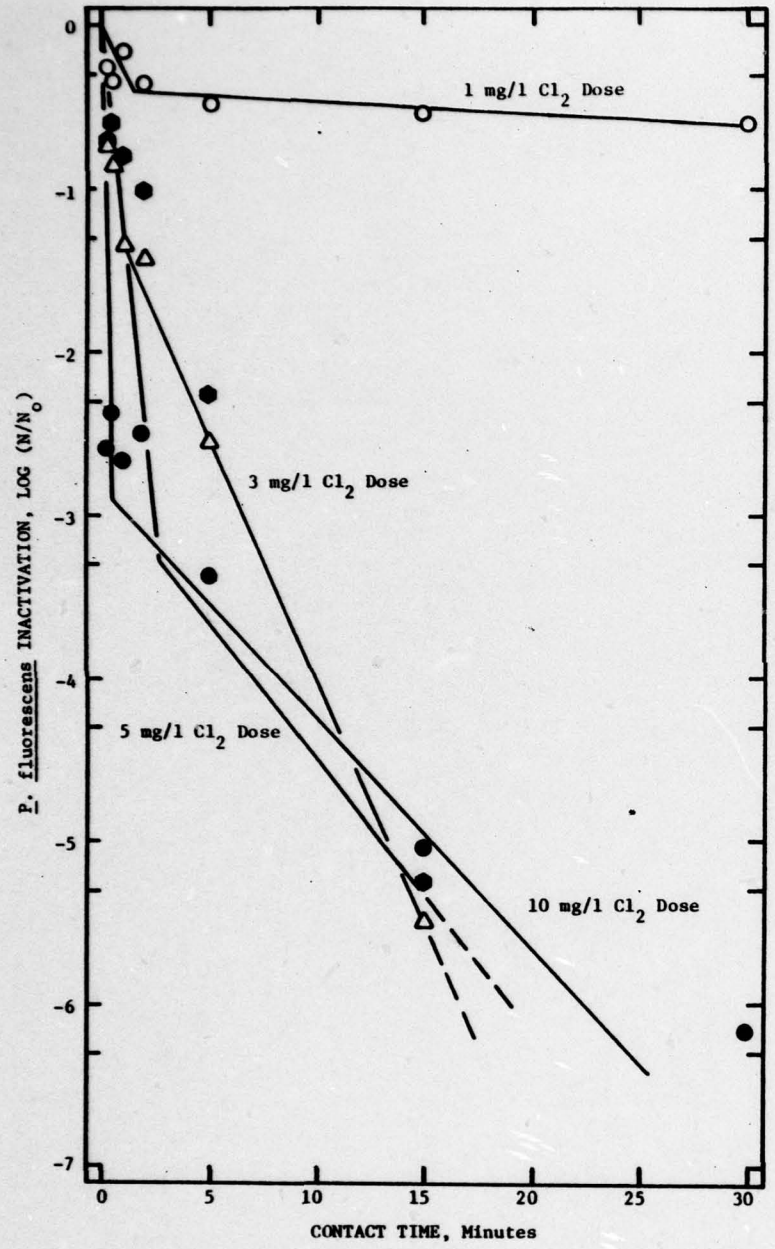


FIGURE 12A. P. fluorescens inactivation as a function of total chlorine dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

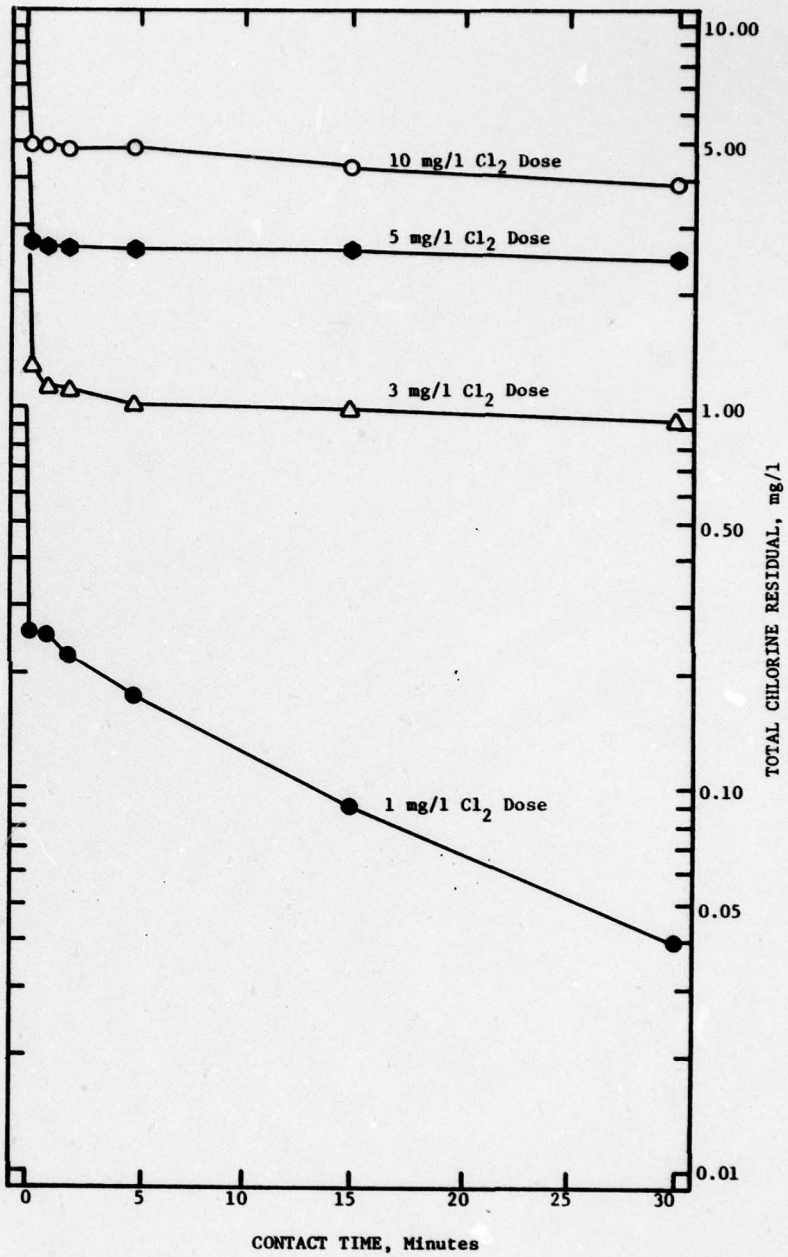


FIGURE 12B. Total chlorine residual as Cl$_2$ as a function of total chlorine dose and contact time for P. fluorescens disinfection studies.

approximately seven logs of inactivation after 30 minutes of contact time was achieved using a chlorine dose of 10 mg/l. The amount of inactivation achieved during the first phase, as shown on Figure 12B, was a direct function of chlorine dose. However, the amount of inactivation demonstrated during the second phase at contact times of about 15 minutes or greater was an inverse function of chlorine dose for the chlorine doses of 3, 5, and 10 mg/l. The chlorine residual curves observed during the chlorine disinfection of P. fluorescens (Figure 12B) are typically biphasic with the initial phases persisting less than 0.5 minutes and characterized by a very rapid chlorine demand rate. The second phases of the chlorine residual curves have a greatly reduced chlorine demand rate relative to the first stages, and the terminal chlorine residuals are a direct function of the chlorine doses varying from approximately 0.04 mg/l for the 1 mg/l chlorine dose to 4 mg/l for the 10 mg/l chlorine dose. Free chlorine was present during the first phase becoming non-detectable within 15 seconds.

Figure 13A shows the inactivation efficiency of iodine for E. coli. The curves are typically biphasic with relatively little inactivation achieved after one minute contact time. Slightly greater than one log of inactivation was achieved for an iodine dose of 1 mg/l and 30 minutes contact time. Nearly seven logs of inactivation were achieved using an iodine dose of 10 mg/l after only five minutes of contact time. Results obtained for iodine doses of 3 and 5 mg/l were intermediate to those described above for 1 and 10 mg/l iodine doses.

As shown on Figure 13B, no iodine residual was detected at 0.25 minutes contact time for 1 mg/l iodine dose, no iodine residual was detected at one minute contact time for the 3 mg/l iodine dose, and no iodine residual was detected at five minutes contact time for the 5 mg/l iodine dose. The curve

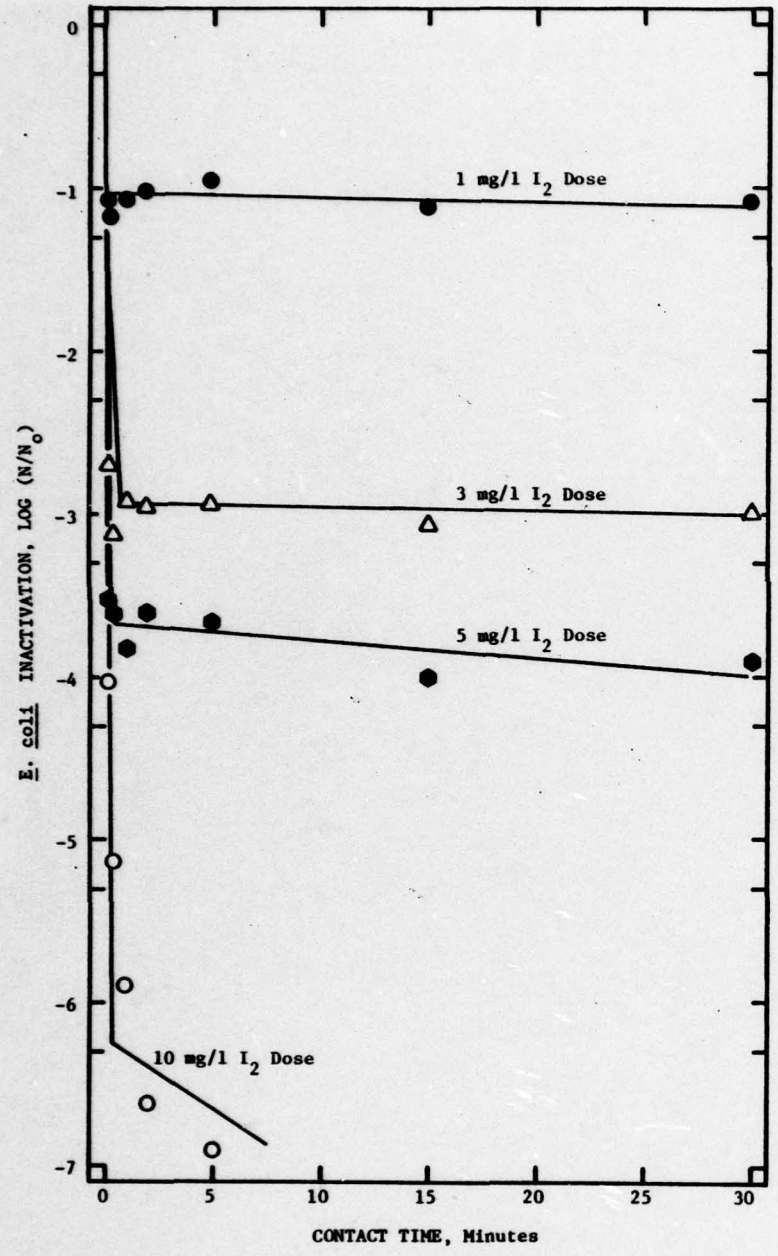


FIGURE 13A. E. coli inactivation as a function of iodine dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

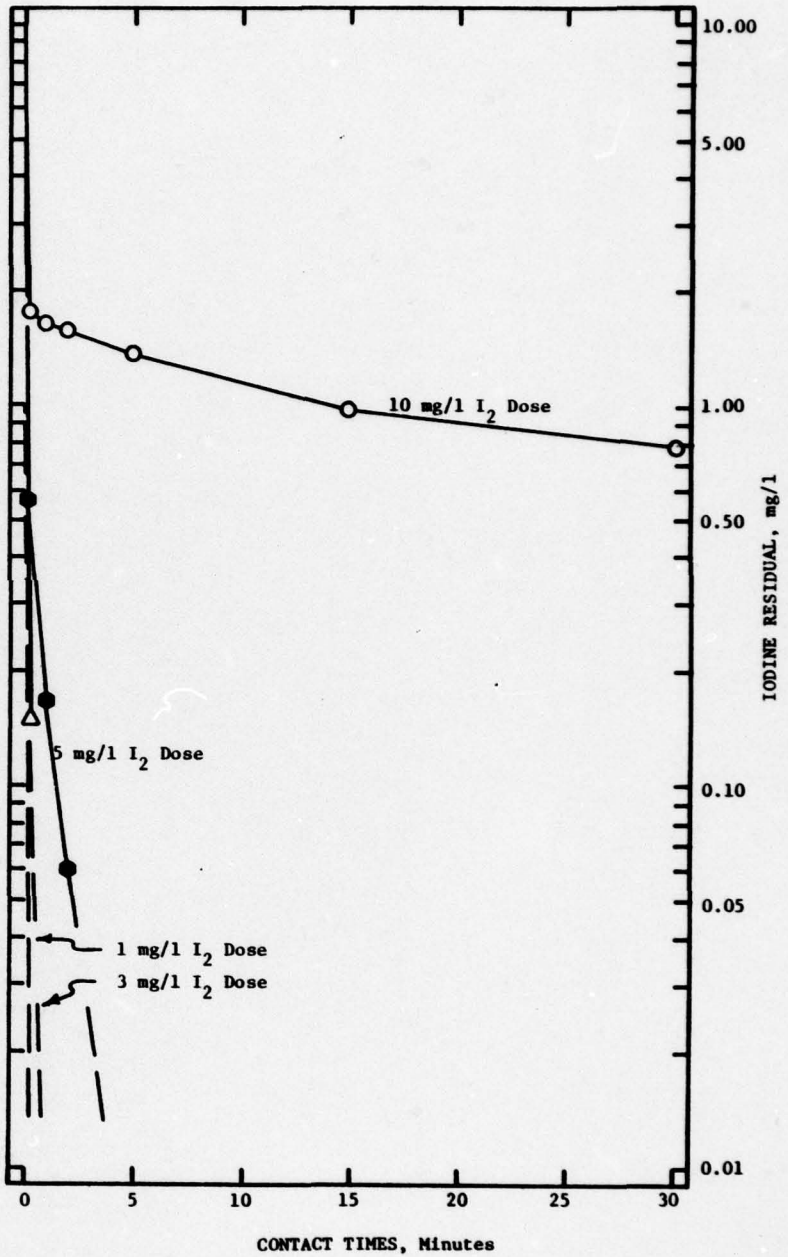


FIGURE 13B. Iodine residual as I_2 as a function of iodine dose and contact time for E. coli disinfection studies.

for the 10 mg/l iodine dose is typically biphasic and shows an iodine residual of approximately 0.8 mg/l after 30 minutes contact time.

As shown on Figure 14A, an iodine bromide dose of 1 mg/l produced an E. coli inactivation of approximately 0.3 log. Iodine bromide doses of 3 and 5 mg/l yielded E. coli inactivations of approximately 3.5 logs and 4.8 logs, respectively. And, using an iodine bromide dose of 10 mg/l, the E. coli inactivation exceeded 6.5 logs after five minutes contact time. All inactivation curves were typically biphasic.

The iodine bromide residuals shown in Figure 14B were typically biphasic for iodine bromide doses of 5 and 10 mg/l, with residuals of 0.02 and 0.7 mg/l, respectively, after 30 minutes contact time. No iodine bromide residual was detectable for doses of 1 and 3 mg/l after contact times of 0.25 minutes and one minute, respectively.

Figure 15A shows results obtained when inactivating E. coli using chlorine as the disinfectant. The inactivation curve for the 1 mg/l chlorine dose was monophasic. Whereas, the inactivation curves for chlorine doses of 3, 5, and 10 mg/l were typically biphasic. For the 1 mg/l chlorine dose, only one log of inactivation was achieved after 30 minutes contact time. However, greater than six logs of E. coli inactivation were achieved for chlorine doses of 5 and 10 mg/l and contact times of 30 and 0.25 minutes, respectively. Approximately 5.2 logs of E. coli inactivation were achieved using a 3 mg/l chlorine dose.

The residuals observed when inactivating E. coli using chlorine as the disinfectant are shown on Figure 15B. The curves are typically biphasic and the total chlorine residuals after 30 minutes contact time vary from 0.01 mg/l for a chlorine dose of 1 mg/l to 3 mg/l for a chlorine dose of 10 mg/l. Free

chlorine persisted in measurable concentrations for 15 seconds at a dose of 10 mg/l.

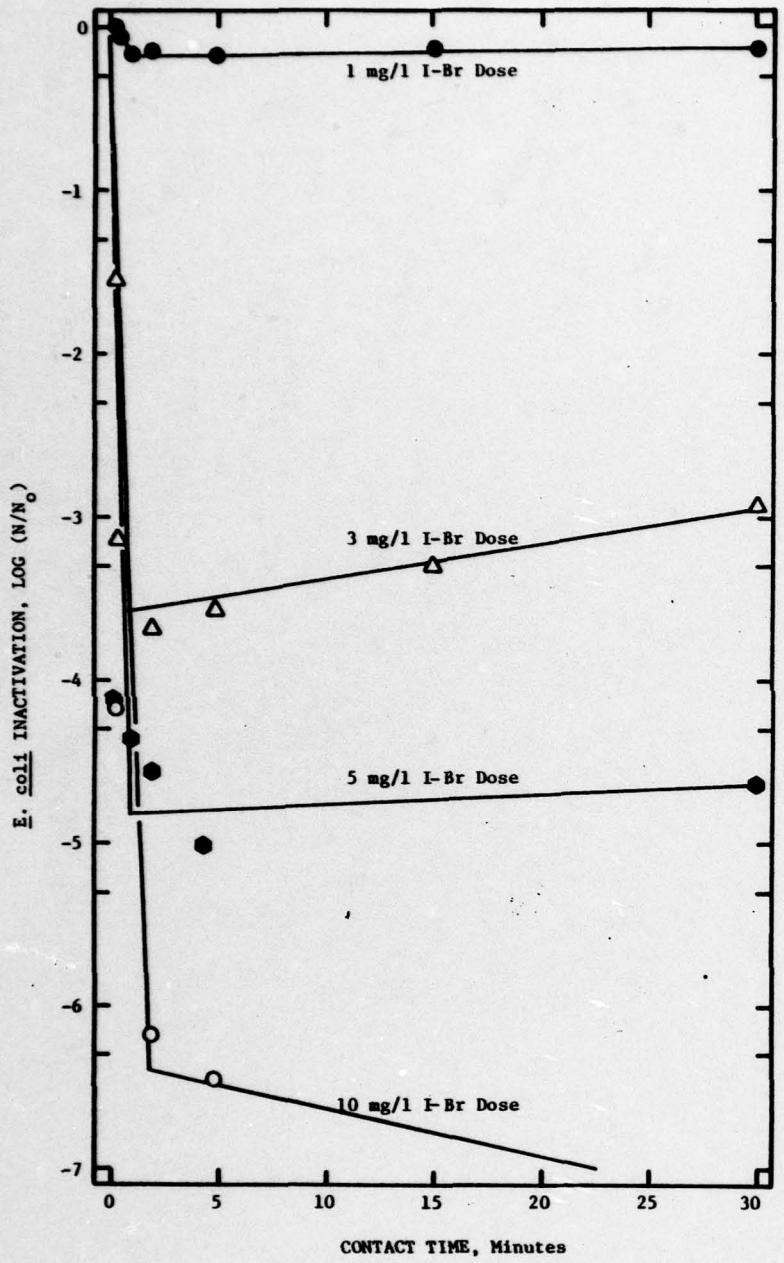


FIGURE 14A. E. coli inactivation as a function of iodine bromide dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

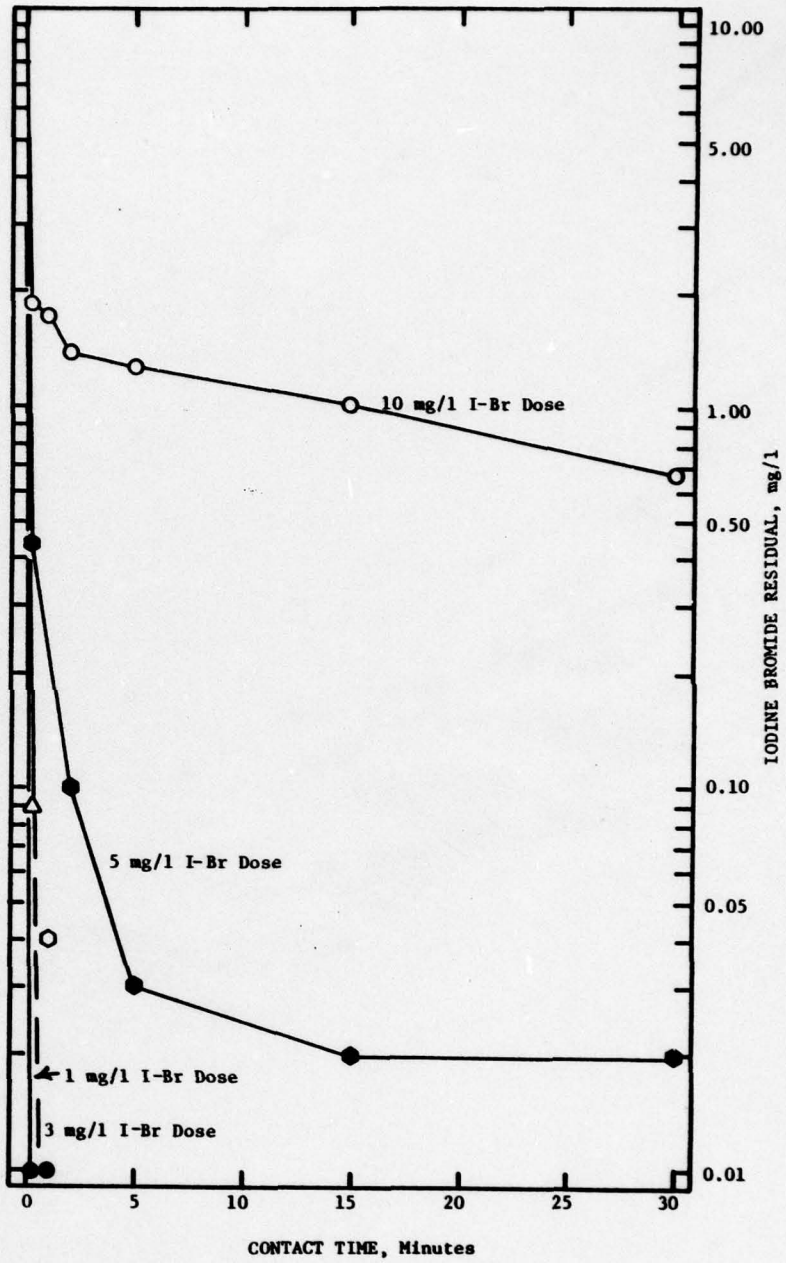


FIGURE 14B. Iodine bromide residual as I_2 as a function of iodine bromide dose and contact time for E. coli disinfection studies.

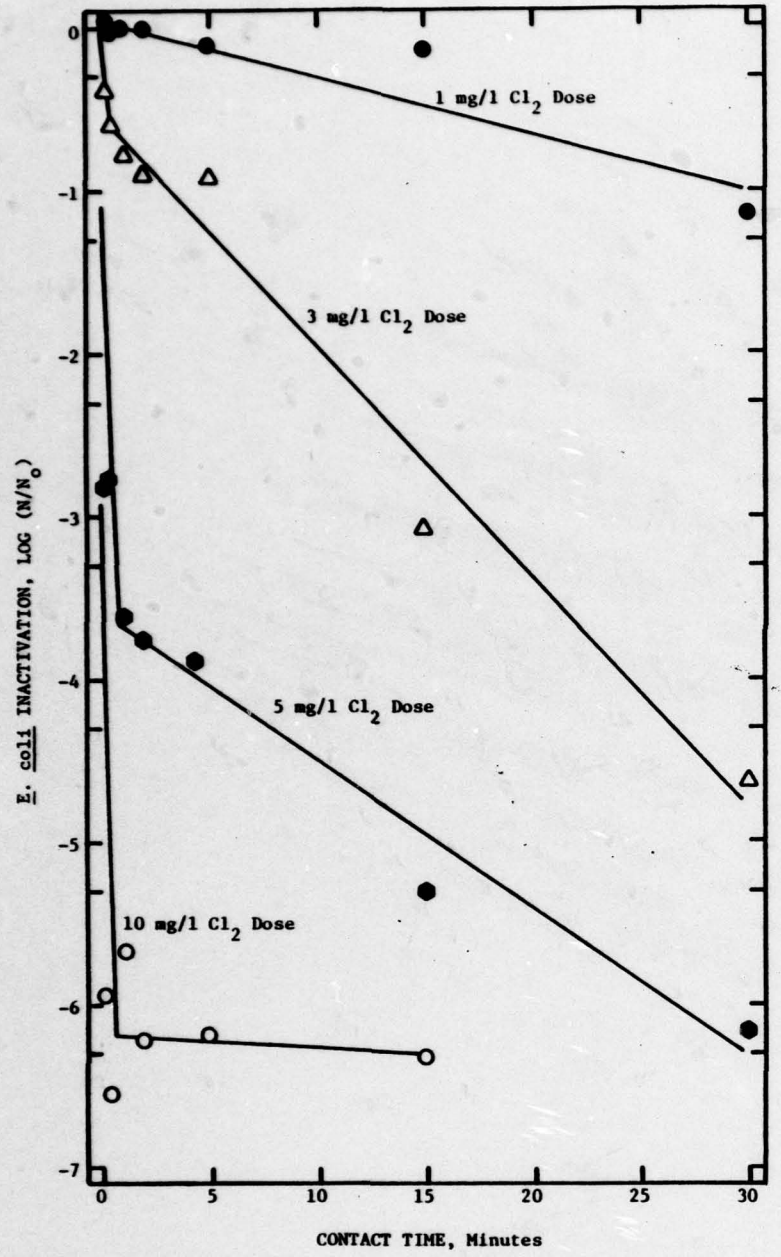


FIGURE 15A. E. coli inactivation as a function of total chlorine dose and contact time at 25 ± 1C and pH 6.9 ± 0.2.

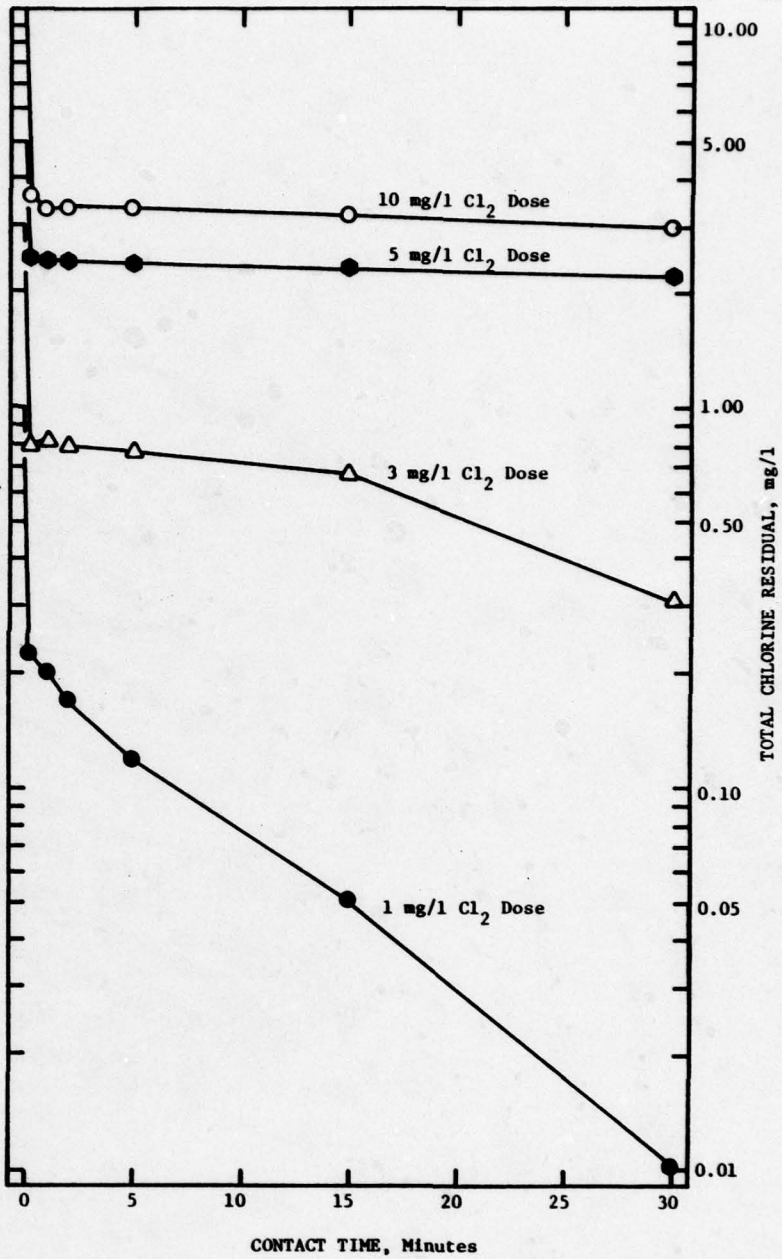


FIGURE 15B. Total chlorine residual as Cl_2 as a function of total chlorine dose and contact time for E. coli disinfection studies.

DISCUSSION

Membrane Studies. The immersion of polyamide membrane in 1, 5, and 50 mg/l of a buffered iodine solution (pH=6.9) for 2000 hours results in considerable absorption of iodine, this phenomona being predominant at high iodine concentrations. Whether this absorption of iodine leads to major structural changes and/or degradation of the membrane is not clear from the immersion studies alone. Analysis of the SEM micrographs do indeed indicate that at high (50 mg/l) concentrations of iodine, structural deterioration of the membrane occurred. At the lower concentrations of 1 and 5 mg/l, evidence for structural change from degradation by the iodine was apparent after long immersion times (Table 2). Length of service of the polyamide membrane if exposed to 1 or 5 mg/l cannot be estimated since flux characteristics may influence the membrane lifetime and degradation characteristics by the iodine. Nevertheless, the 5 mg/l concentration of iodine was judged to be a satisfactory disinfectant with regard to the test organisms examined and will cause relatively minor structural changes on the membrane if contact time is less than an hour.

The surface appearance of the polyamide membrane at time zero or immersed only in the buffer solution is similar to the surface described by Kesting for polycellulose acetate membranes (1). The nodules observed on the polyamide membrane surface appear to be similar in shape (ellipsoid) to those observed on the polycellulose membrane surface. However, the observed nodule size is approximately 5- to 6-fold larger for the polyamide membrane. The consistancy of these dimensions in all SEM's prepared supports the idea that the nodules are characteristic of RO-membranes. As time of immersion in the

iodine disinfectant solutions increases, the changes observed in nodule size and appearance parallel the apparent structural deterioration of the membrane surface. All iodine concentrations exhibited the same progression of effects, albeit at different lengths of time, ranging from initial nodule expansion to nodule shrinkage, to appearance of "holes" or structural anomalies in the surface.

Schultz and Asunmaa have analyzed polycellulose acetate membranes and proposed that clusters of bulk water are in a highly hydrogen-bonded structure with the membrane surface and form a hydration sheath of appreciable thickness (13). If such a hydration sheath is formed, the solubility of iodine or other salts and solutes should be considerably lowered compared to bulk water (13). Consequently, protection of the surface would be achieved. High concentrations of the iodine disinfectant could disrupt this hydration sheath making the membrane surface susceptible to attack. The observed changes in the polyamide membrane nodules could be due to alteration in this hydration sheath leading to the changes in surface structure.

Disinfection Studies. It was not possible to compare equal halogen doses on a weight basis since approximately 80 percent of the free chlorine was initially present as hypochlorous acid, the remainder being present as hypochlorite ion. The free chlorine persisted in solution for not more than 15 seconds following the initial introduction of the chlorine solution for each test organism. After 15 seconds, a reaction has taken place with nitrogenous compounds. Such compounds, introduced initially with the organism, result in the formation of chloramines. Therefore, any chlorine induced bacterial inactivation occurring during the second stage of the disinfectant curve must occur in the presence of or be due to chloramines.

The biphasic curves, or "L-shaped curves," were present for all test disinfectants. An initial rapid halogen demand (Figures 7 through 15) concurrent with rapid initial bacterial inactivation occurred during the first stage of the curve for all test halogens. The decreased rate of inactivation during the second stage of the curves can result from: (a) conversion of the biocidal species to a form having greatly decremented disinfection properties; (b) protection of the remaining viable organisms by inclusion in solids in which case the diffusion of the disinfectant through the surrounding solids may become the disinfection rate limiting parameter; or, (c) survival of bacteria more resistant to the disinfectant than those initially inactivated. Of the three foregoing hypotheses, the first seems least probable. While the chlorine does form less biocidal species (chloramines predominate during the second stage of the curves), iodine does not react to form iodamines in detectable concentrations at the test pH. Presumably the biocidal iodine species predominating during both stages of the curves is iodine as I_2.

Most of the inactivation achieved using chlorine occurred during the first stage while free chlorine remained in the disinfection system. While second stage chlorine inactivation was relatively insignificant, the study did not evaluate the biocidal properties of the chloramines when they are introduced initially as the primary disinfectant. Since the chloramines have lower oxidation potentials relative to the elemental halogens, or their hypohalic acid analogs, they may not be as deleterious to the RO membrane as iodine or free chlorine. Further, chloramines are considerably more stable than the bromamines which have such a short half-life in solution. It may be difficult to maintain desired bromamine residuals throughout a RO membrane system, even if the bromamines have little interaction with RO membrane material.

The inactivation of P. fluorescens (Figure 12A), exhibits an inverse relationship between chlorine dose and the inactivation rate during the second stage. Contact times of 15 minutes or greater and chlorine doses greater than 1 mg/l demonstrate an inactivation which varies inversely as a function of chlorine dose. This anomaly was not observed during the initial disinfection stage and is not explained by the chlorine residual data which is typically biphasic (Figure 12B). However, comparison of the chlorine residual data after 30 minutes contact time, as shown in Table 3, reveals that a higher total chlorine residual persisted for the P. fluorescens experiments relative to the P. aeruginosa, and E. coli experiments for chlorine doses of 3, 5, and 10 mg/l. This difference in residual could be explained as being due to a greater chlorine demand in the P. fluorescens and E. coli reaction systems. However, comparison of corresponding data for the iodine and iodine bromide does not support the foregoing hypothesis. Alternatively, inactivation of P. fluorescens using chlorine may tend to be dose independent when the biocidal species of chlorine are chloramines.

Table 3. Halogen Residual At 30 Minutes Contact Time For Test Bacterial Organisms

Organism	Halogen Dose, mg/l	Total Halogen Residual, mg/l		
		Iodine	Iodine Bromide	Chlorine
P. aeruginosa	1.0	0.04	0.01	0.21
P. fluorescens	1.0	<0.01	<0.01	0.04
E. coli	1.0	<0.01	<0.01	0.01
P. aeruginosa	3.0	<0.01	<0.01	0.80
P. fluorescens	3.0	0.02	<0.01	0.92
E. coli	3.0	<0.01	<0.01	0.50
P. aeruginosa	5.0	0.40	1.10	2.11
P. fluorescens	5.0	0.39	0.04	2.45
E. coli	5.0	<0.01	0.02	2.19
P. aeruginosa	10.0	1.65	1.72	2.11
P. fluorescens	10.0	1.80	1.49	3.86
E. coli	10.0	0.78	0.67	2.97

The Ct product curves on Figures 16 through 18 can be interpreted to support the enhanced bactericidal efficiency of both iodine and iodine bromide relative to chlorine. At a comparably low Ct product (∿30 mg-sec/1) approximately three logs of inactivation for E. coli were achieved using iodine (Figure 16) and only one log inactivation with chlorine (Figure 18). Therefore, iodine appears to be a much better disinfectant on a weight basis for E. coli in a system characterized by an initial and appreciable halogen demand. Similar conclusions can be made for the Ct products for P. aeruginosa and P. fluorescens. However, an artifact exists in the Ct product data on Figures 16 through 18. The initial high iodine demand relative to the chlorine demand for a given halogen dose, as shown by the 1-minute demand on Figures 19 through 21, results in an initial lower iodine residual. Consequently, lower Ct products are observed for iodine even through the overall bacterial inactivation after 30 minutes contact time may be similar for both disinfectants.

Iodine bromide reacts in aqueous solution to form iodine in equilibrium with the parent compound as shown by equation 6. Therefore, iodine may be the

$$2\ IBr \rightleftharpoons I_2 + 2\ Br^- \qquad (6)$$

biocidal agent present when iodine bromide is used as a disinfecting agent. Consequently, the statements made relative to the use of iodine as a disinfecting agent may also apply to iodine bromide.

Differences between these two compounds exist in the relative health hazards and operational problems associated with their storage and handling. However, inactivation rates for iodine bromide tend to be slower than for iodine in similar test configurations.

Pseudomonas species investigated in this study tend to be more resistant than E. coli to iodine, iodine bromide or chlorine with this characteristic becoming more pronounced as the Ct product values increase above 10mg-sec/1.

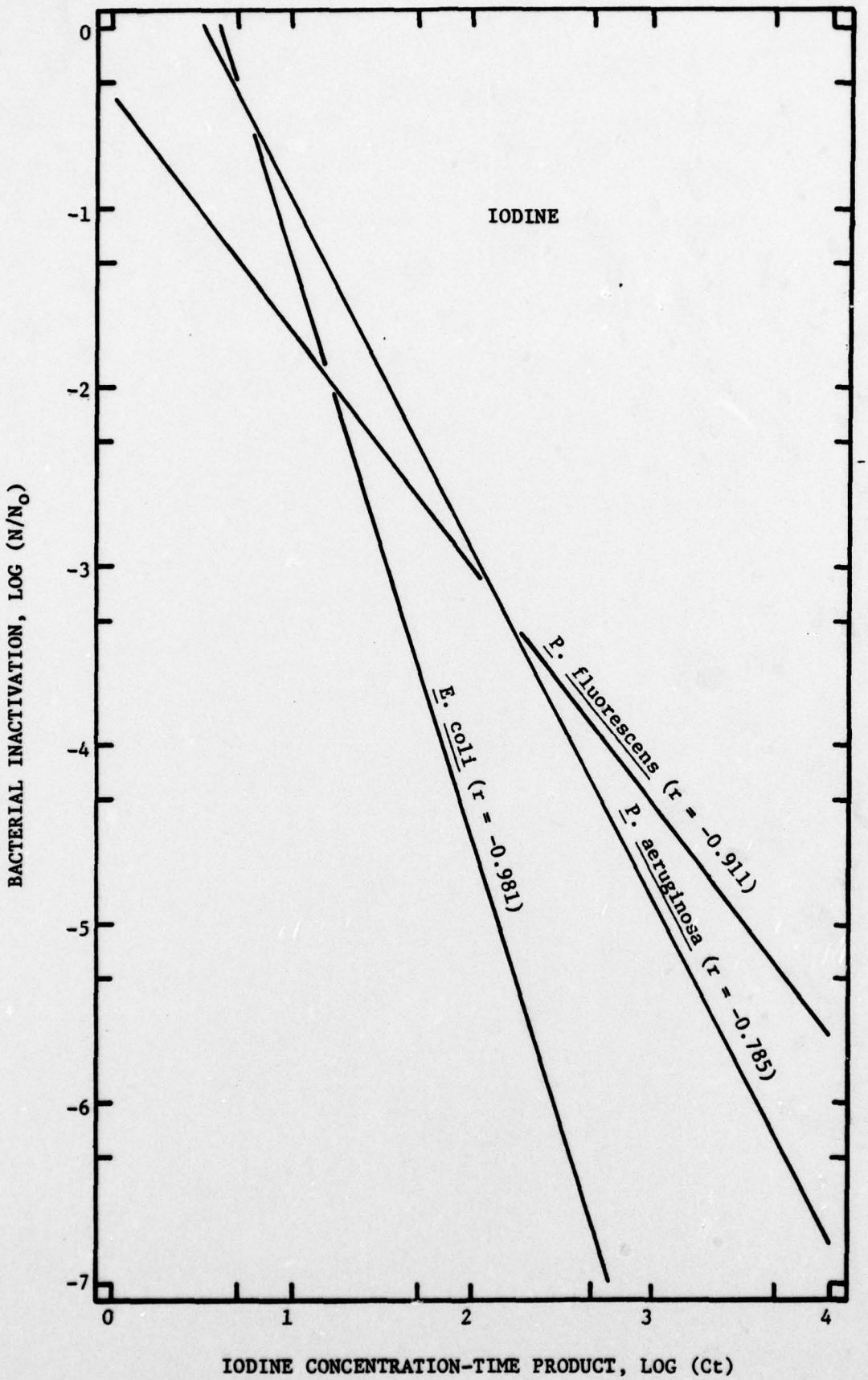


FIGURE 16. P. aeruginosa, P. fluorescens, and E. coli inactivation for iodine disinfection studies as function of concentration-time product (mg-sec/l).

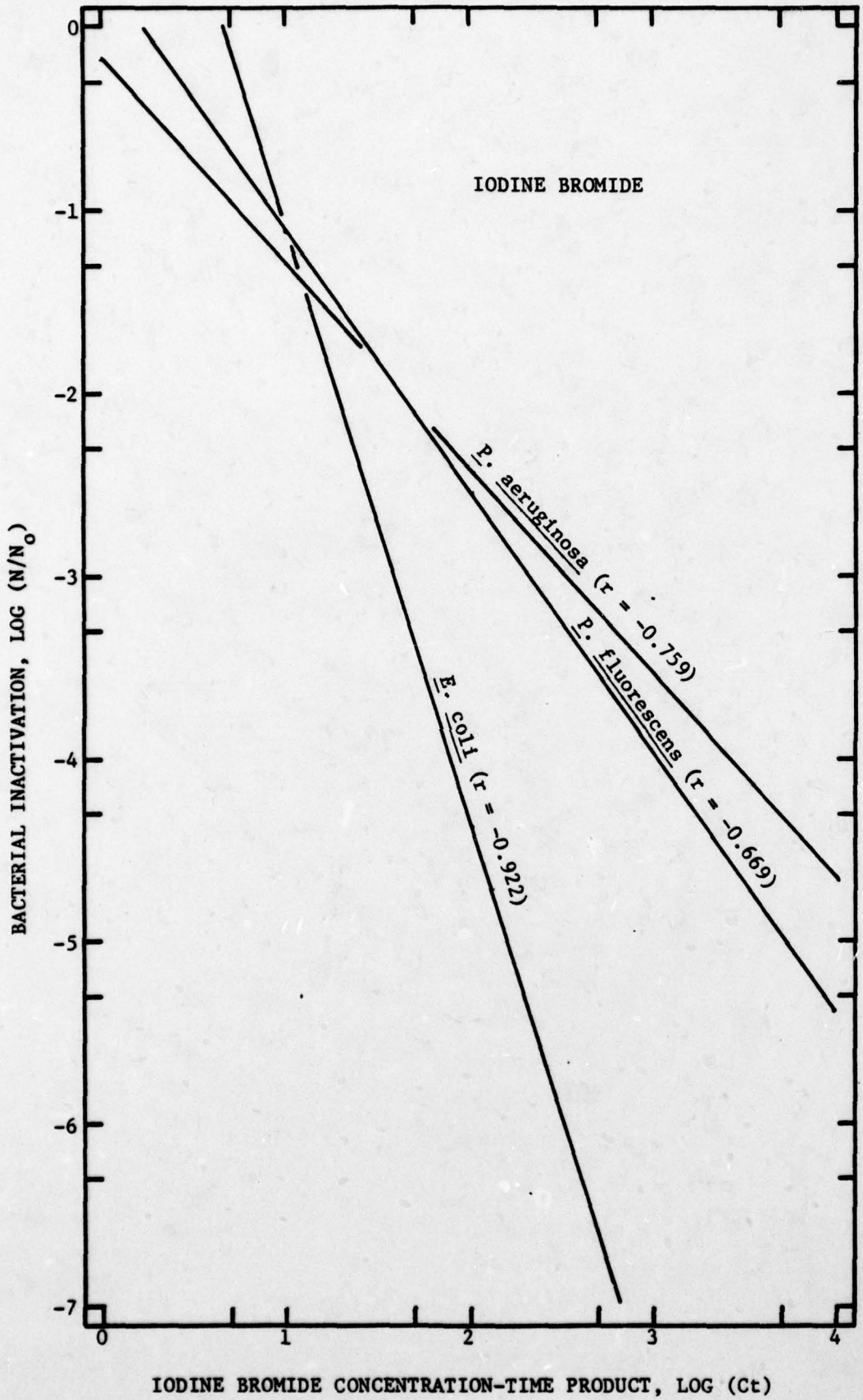


FIGURE 17. P. aeruginosa, P. fluorescens, and E. coli inactivation for iodine bromide studies as function of concentration-time product (mg/sec/l).

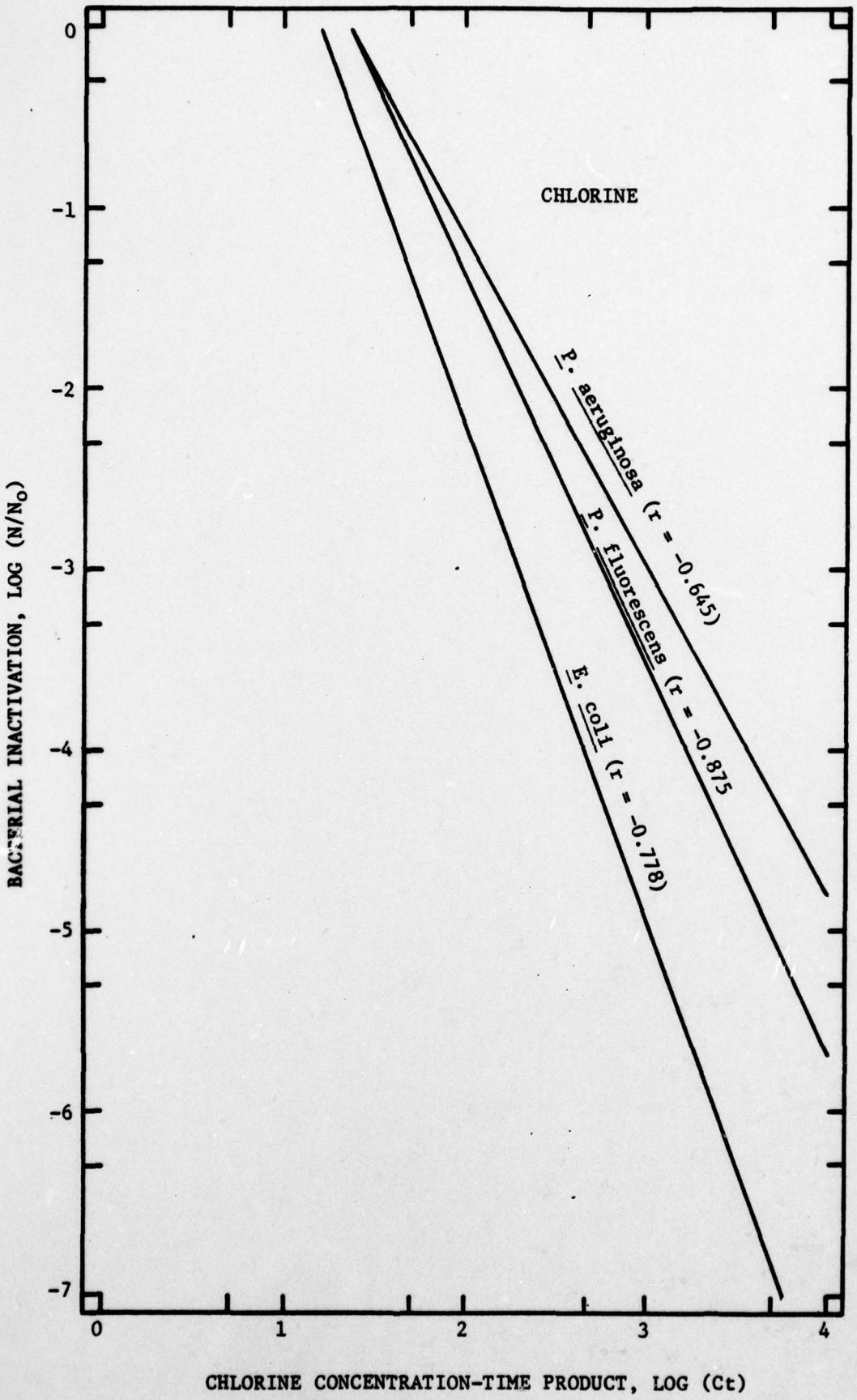


FIGURE 18. P. aeruginosa, P. fluorescens, and E. coli inactivation for chlorine disinfection studies as function of concentration-time product (mg-sec/l).

Therefore, iodine doses based on E. coli inactivation would be too conservative for RO membrane maintenance, especially if the water applied to the RO membrane contains substances susceptible to iodine demand or if the iodine demand exerted by the membrane itself is not considered. Furthermore, while iodine doses of 5 mg/l resulted in approximately four logs of inactivation for all test organisms after 30 minutes contact time, appreciable numbers of test organisms (8.4×10^4 to 7.9×10^6 colony forming units per litre) survived for 30 minutes after being subjected to a 5 mg/l iodine dose.

The disinfection studies were carried out in a batch system, and therefore the hydraulic characteristics depart significantly from the continuous flow mode under which a reverse osmosis unit is operated. However, the disinfected liquid slug in the batch mixer is analogous to a liquid slug entering a RO membrane after being subjected to a chemical disinfectant such as iodine. While the disinfected liquid slug entering the RO membrane would be both partitioned and subjected to backmixing in the membrane, addition of disinfectant within the RO membrane is not feasible. If the disinfectant is introduced immediately upstream of the RO membrane, the average age of any disinfectant residual in the effluent should correspond to the average residence time of the liquid slug in the membrane. Therefore, as iodine is subjected to demand downstream in the RO membrane, both by the membrane material and demand producing constituents on the membrane or in the feed-water, the chemical disinfectant residual is dissipated with a concurrent decrease of stress on surviving organisms. Thus, sufficiently high iodine doses may be required to produce a biocidal residual throughout the membrane resulting in significant membrane deterioration. This effect would be most predominant at the upstream end of the membrane.

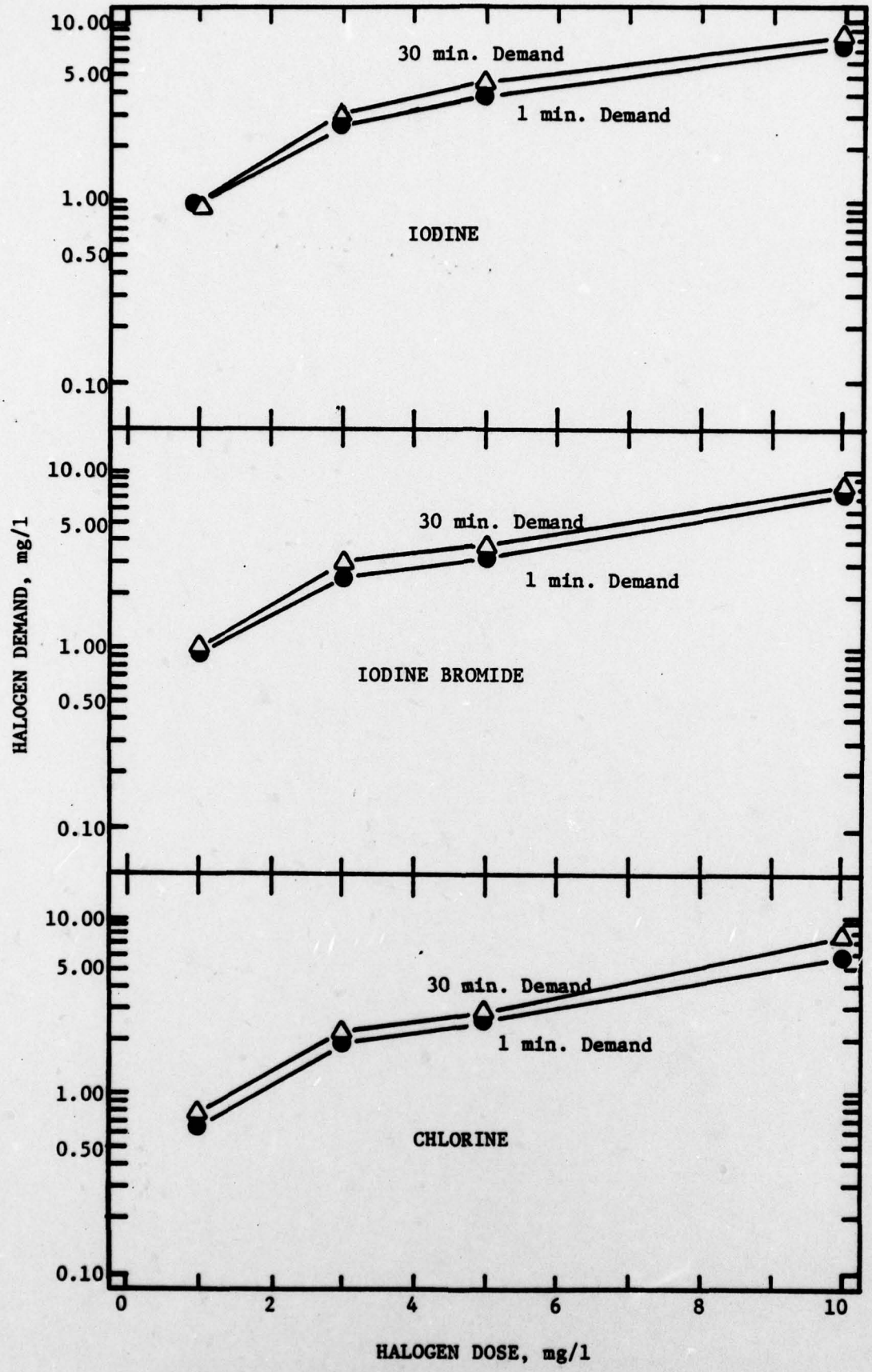


FIGURE 19. Iodine, iodine bromide, and chlorine demand as a function of halogen dose and 1 and 30 minutes contact time 25 ± 1C and pH 6.9 ± 0.2 for P. aeruginosa disinfection studies.

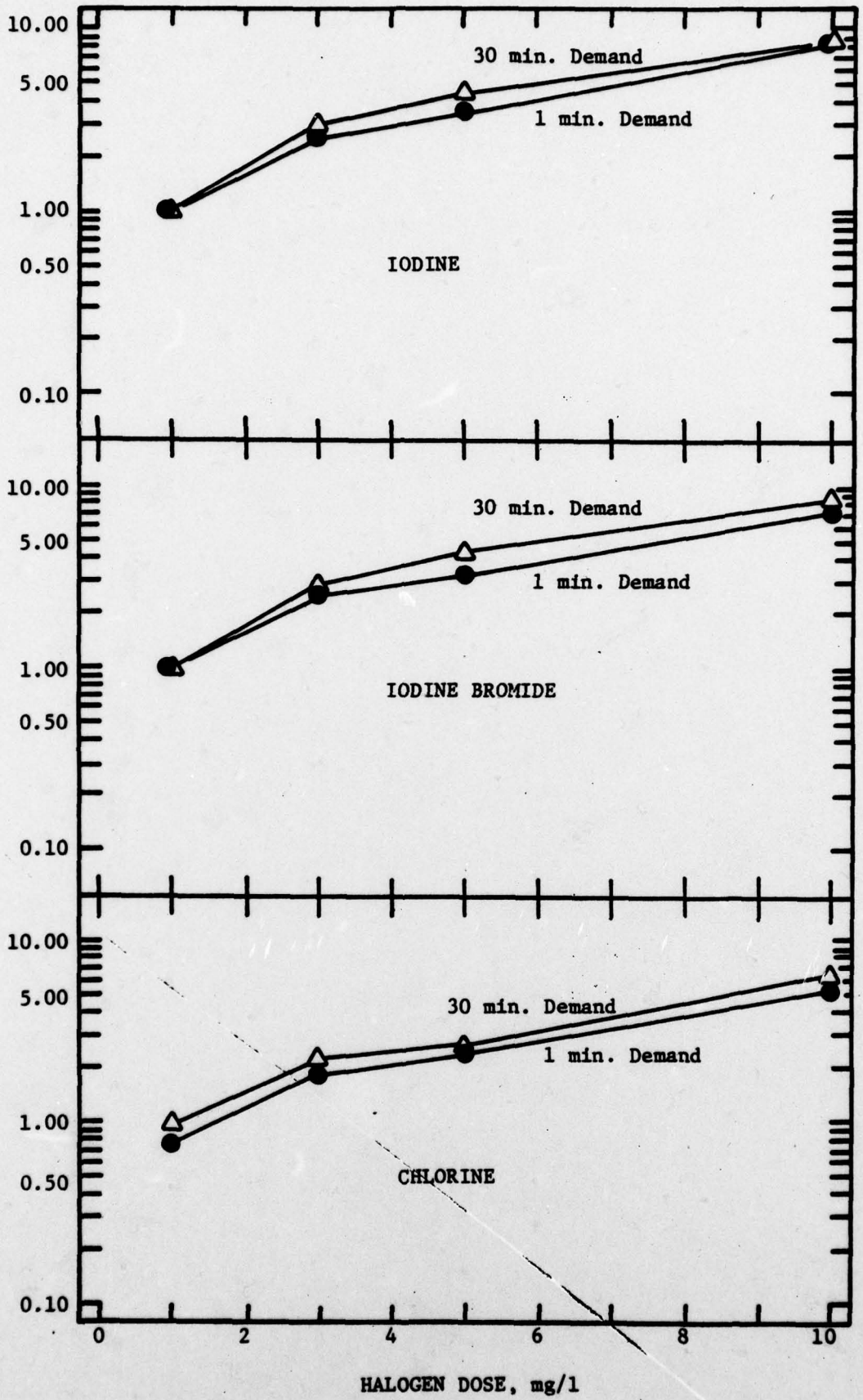


FIGURE 20. Iodine, iodine bromide, and chlorine demand as function of halogen dose and 1 and 30 minutes contact time 25 ± 1C and pH 6.9 ± 0.2 for P. fluorescens disinfection studies.

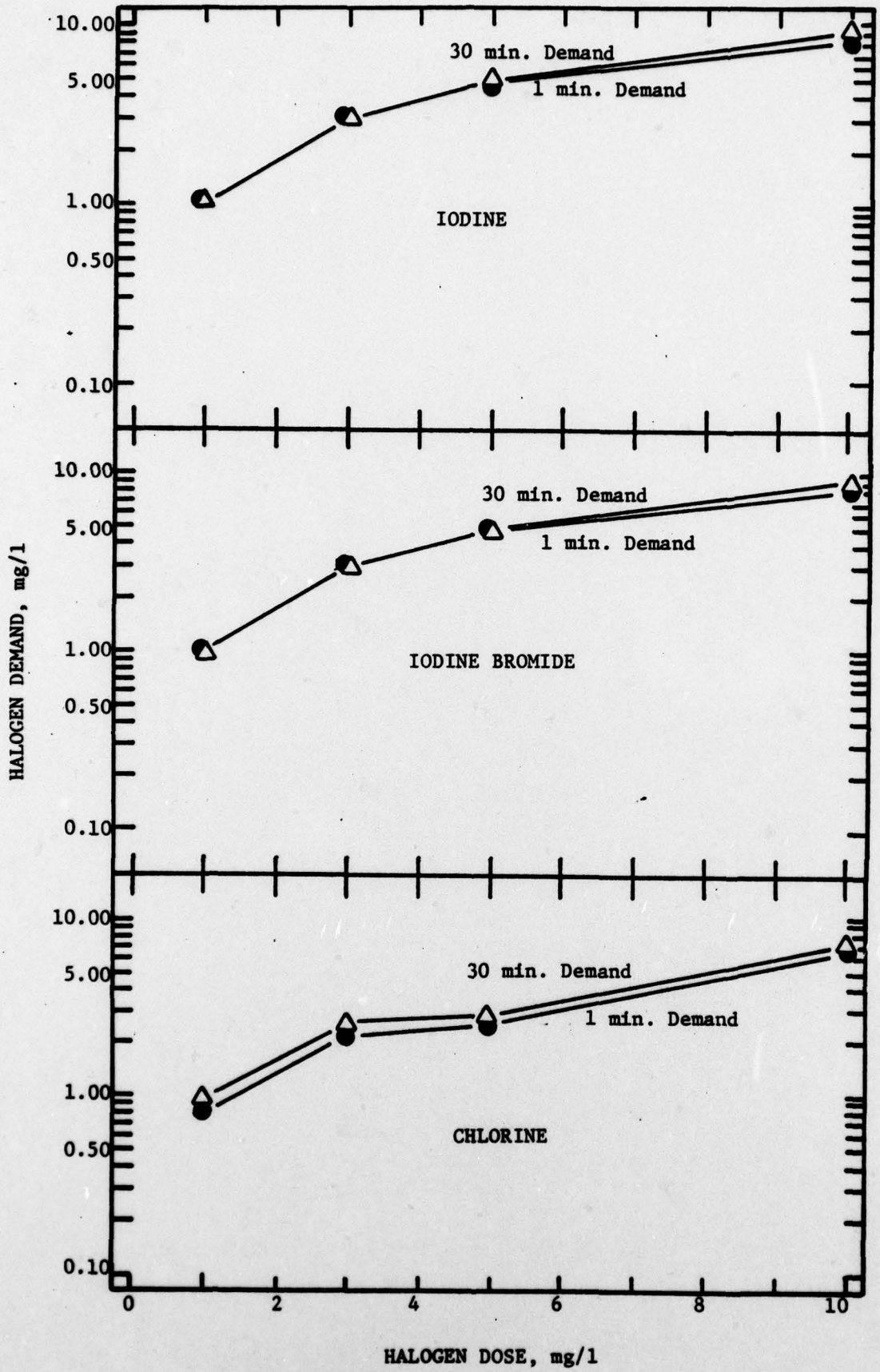


FIGURE 21. Iodine, iodine bromide, and chlorine demand as a function of halogen dose and 1 and 30 minutes contact time at 25 ± 1C and pH 6.9 ± 0.2 for E. coli disinfection studies.

CONCLUSIONS

1. Significant polyamide membrane deterioration occurs which is discernible by the appearance of holes in the membrane after 45 days of immersion in 1 and 5 mg/l buffered iodine solutions and after 5 days immersion in a 50 mg/l buffered iodine solution.
2. *P. aeruginosa*, *P. fluorescens* and *E. coli* are successfully recovered from an RO-membrane which had been operated using Potomac River water.
3. *P. aeruginosa* and *P. fluorescens* are more resistant than was *E. coli* to inactivation using either iodine, iodine bromide, or chlorine as the disinfectant.
4. The half-life of bromamines is so short that their use as a bactericidal or bacteriostatic agent for membrane maintenance may not be feasible.
5. Iodine and iodine bromide are excellent bactericidal agents. Their bactericidal efficiency appears to be comparable to the chlorine disinfection system used in this study.
6. The chlorine disinfection system used in this study was generally characterized by the presence of free chlorine for less than 0.25 minutes followed by persisting chloramine residual. The chloramines, as employed, did not demonstrate appreciable bactericidal efficiency.
7. The significant bacterial inactivation achieved during the first stage of chlorine disinfection presumably in the presence of free chlorine, was observed during the first inactivation stages using iodine and iodine bromide as disinfectants also. Further, these latter disinfectants were characterized by an initial rapid bacterial inactivation followed by a much slower rate of inactivation during the second stage when iodine (as I_2) was presumably the prime biocidal agent remaining in the solution.

8. The biphasic curves may result from initial inactivation of the susceptible organisms rather than a change in the biocidal species.

APPENDIX A

IMMERSION STUDIES OF POLYAMIDE MEMBRANE STRIPS IN BUFFERED IODINE SOLUTIONS (pH=6.9; 0.01M phosphate; 27C) IN GLASS REACTION VESSELS WITH A CONSTANT HEAD SPACE

The glass tanks were changed at the indicated times by transferal of the membrane strip into another tank of the same initial concentration. The amount absorbed by the membrane (corrected for head space volatilization from control measurements) is given in column three and the calculated absorption rate in mg/l per hour is given in column four. Excursion from the initial desired iodine concentration was ±5% throughout the 2000-hour study.

Table A-1. Initial iodine concentration of 1 mg/l

Table A-2. Initial iodine concentration of 5 mg/l

Table A-3. Initial iodine concentration of 50 mg/l

Table A-1

Tank Change Number	Elapsed Time (Hours)	Iodine Concentration Total Absorbed in mg/l	Iodine Concentration Absorption Rate in mg/l/hr.
1	1	.0308	.0308
2	4	.135	.0348
3	8	.218	.0207
4	12	.281	.0159
5	14	.319	.0189
6	19.2	.394	.0145
7	24	.453	.0122
8	29	.498	.0089
9	35	.768	.0451
10	43	.824	.0070
11	48	.842	.0036
12	56	.867	.0031
13	68	.976	.0091
14	73.5	1.12	.0256
15	80.5	1.18	.0097
16	92.5	1.27	.0073
17	99	1.39	.0187
18	107	1.56	.0207
19	116	1.71	.0162
20	124	1.84	.0171
21	131	1.97	.0179
22	140	2.04	.0085
23	148.3	2.08	.0043
24	158	2.17	.0092
25	172	2.20	.0020
26	184	2.27	.0058
27	212	2.38	.0042
28	222.5	2.47	.0079
29	240	2.58	.0065
30	263.5	2.73	.0062
31	285	2.83	.0048
32	309	2.91	.0033
33	334	2.98	.0028
34	360	3.17	.0071
35	381	3.28	.0053
36	409	3.44	.0057
37	432	3.57	.0058
38	459	3.64	.0023
39	483	3.73	.0038
40	507	3.83	.0044
41	528	3.97	.0066
42	552	4.08	.0046
43	578.5	4.20	.0044
44	604.5	4.30	.0039
45	628.5	4.36	.0023
46	649.5	4.38	.0014

(Table A-1 continued)

Tank Change Number	Elapsed Time (Hours)	Iodine Concentration Total Absorbed in mg/l	Iodine Concentration Absorption Rate in mg/l/hr.
47	679.5	4.48	.0030
48	706.5	4.62	.0054
49	736.5	4.71	.0029
50	769.5	4.81	.0028
51	793.5	4.82	.0005
52	821.5	4.97	.0053
53	849	5.07	.0036
54	882	5.21	.0042
55	911	5.27	.0022
56	941	5.31	.0015
57	977	5.37	.0012
58	1007	5.38	.0005
59	1035	5.44	.0022
60	1059	5.54	.0044
61	1087	5.64	.0036
62	1123	5.69	.0013
63	1151.5	5.70	.0005
64	1181	5.75	.0016
65	1203	5.80	.0021
66	1233	5.87	.0026
67	1271	5.95	.0020
68	1301	6.01	.0018
69	1327	6.14	.0050
70	1362	6.26	.0034
71	1392	6.33	.0023
72	1424	6.42	.0028
73	1457	6.44	.0006
74	1487	6.50	.0021
75	1511	6.57	.0027
76	1539	6.64	.0024
77	1567	6.70	.0022
78	1603	6.81	.0032
79	1639	6.90	.0024
80	1665	6.97	.0026
81	1700	7.04	.0021
82	1729	7.08	.0012
83	1760	7.16	.0026
84	1793.5	7.23	.0021
85	1823	7.30	.0022
86	1851	7.35	.0020
87	1889	7.44	.0024
88	1919	7.53	.0030
89	1949	7.64	.0034
90	1975	7.71	.0029
91	2000	7.78	.0026

Table A-2

Tank Change Number	Elapsed Time (Hours)	Iodine Concentration Total Absorbed in mg/l	Iodine Concentration Absorption Rate in mg/l/hr.
1	.5	.197	.393
2	1	.326	.259
3	2.25	.611	.228
4	4	.758	.0840
5	8	.962	.0511
6	13.5	1.11	.0261
7	24.5	1.37	.0244
8	31	1.51	.0208
9	48	1.94	.0251
10	63	2.15	.0144
11	73	2.14	-.0010
12	93	2.56	.0210
13	118	2.79	.0089
14	145	3.73	.0352
15	155.5	3.98	.0232
16	173.5	4.38	.0222
17	203.5	5.00	.0207
18	222	5.41	.0218
19	242	5.81	.0200
20	267	6.15	.0135
21	292	6.55	.0162
22	316	7.04	.0204
23	342	7.47	.0163
24	366.5	7.98	.0210
25	393.5	8.43	.0165
26	433.5	9.11	.0172
27	465	9.63	.0163
28	506	10.0	.0094
29	535	10.4	.0131
30	579	11.2	.0174
31	608	11.7	.0190
32	648	12.3	.0145
33	680	12.8	.0167
34	721	13.6	.0193
35	758	14.3	.0172
36	800	14.8	.0134
37	845	15.4	.0120
38	878	16.0	.0183
39	912	16.5	.0147
40	940	16.9	.0139
41	969	17.2	.0121
42	1014	17.9	.0149
43	1058	18.6	.0155
44	1091	19.1	.0171
45	1137	19.9	.0163
46	1182	20.4	.0118

(Table A-2 continued)

Tank Change Number	Elapsed Time (Hours)	Iodine Concentration Total Absorbed in mg/l	Iodine Concentration Absorption Rate in mg/l/hr.
47	1226	21.0	.0140
48	1272	21.6	.0115
49	1306	21.9	.0108
50	1351	22.4	.0113
51	1394	22.9	.0109
52	1442	23.5	.0117
53	1481	23.8	.0083
54	1522	24.2	.0103
55	1564	24.8	.0132
56	1598	25.2	.0127
57	1641	25.7	.0111
58	1685	26.3	.0130
59	1729	26.8	.0114
60	1775	27.2	.0090
61	1821	27.7	.0105
62	1858	28.2	.0146
63	1902	28.9	.0150
64	1932	29.2	.0124
65	1975	29.7	.0114
66	2000	30.2	.0174

Table A-3

Tank Change Number	Elapsed Time (Hours)	Iodine Concentration Total Absorbed in mg/l	Iodine Concentration Absorption Rate in mg/l/hr.
1	.5	3.13	6.26
2	1	7.69	9.12
3	32.5	12.6	.155
4	99	17.7	.077
5	223	21.4	.030
6	369	24.9	.024
7	492.5	27.7	.022
8	612.5	29.6	.015
9	771	32.7	.019
10	915	36.4	.025
11	1033	38.5	.017
12	1180	41.5	.020
13	1325	43.4	.013
14	1493	47.1	.021
15	1615	49.3	.018
16	1760	52.8	.023
17	1875	56.1	.029
18	2000	59.1	.024

APPENDIX B

SCANNING ELECTRON MICROGRAPHS OF POLYAMIDE MEMBRANE STRIPS IMMERSED IN BUFFERED IODINE SOLUTIONS (pH=6.9; 0.01M Phosphate: 27C) IN GLASS REACTION VESSELS

The membrane strips were cut and prepared for SEM analysis as described in EXPERIMENTAL. Both the front and back of the membrane were examined at three magnifications (1,400X; 7,000X; 14,000X).

Figures B1-12. Iodine concentration of zero; buffer alone.

Figures B13-B17. Iodine concentration of 1 mg/1.

Figures B18-B24. Iodine concentration of 5 mg/1.

Figures B25-B33. Iodine concentration of 50 mg/1.

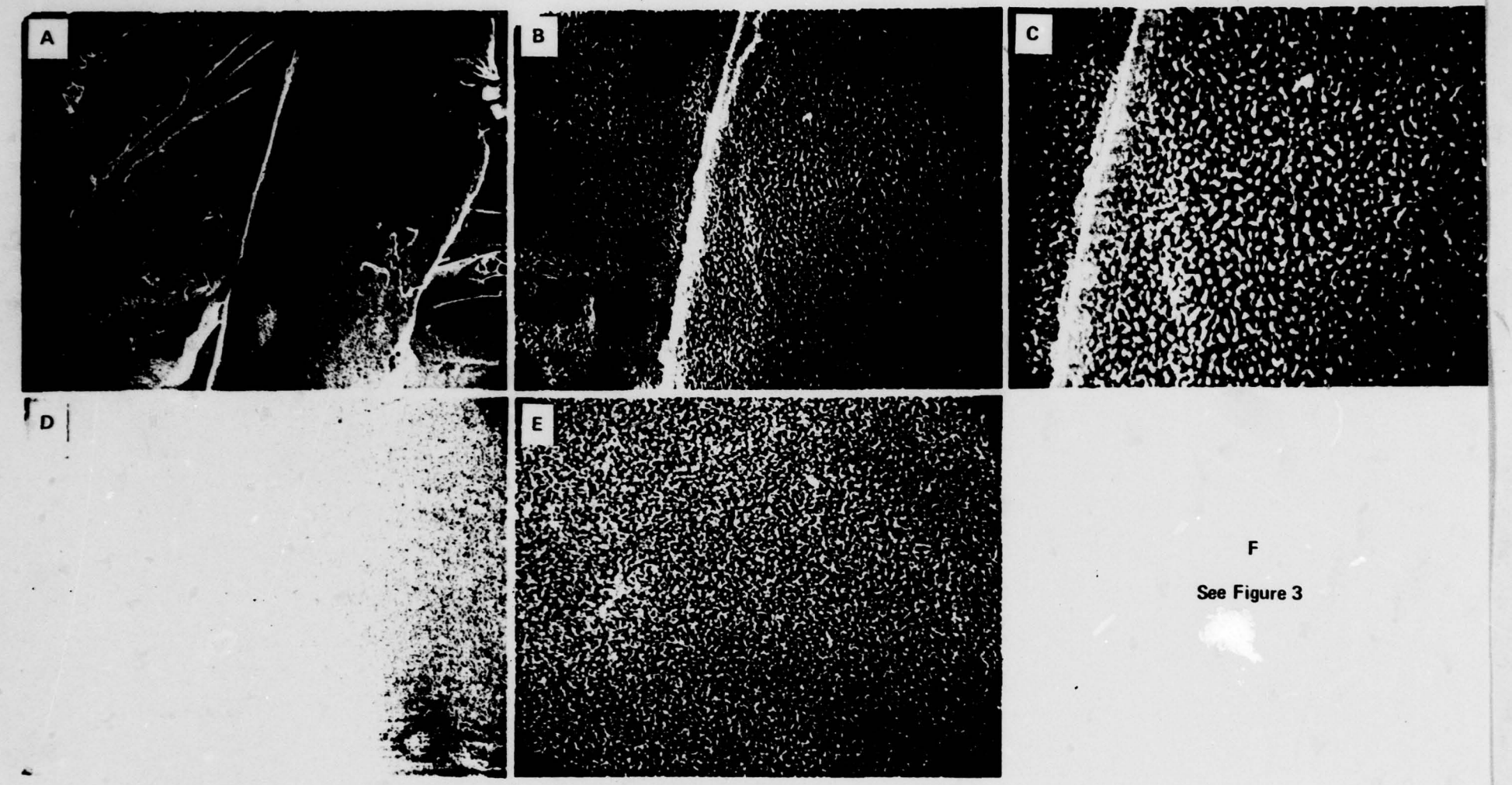


Figure B1. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 0 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

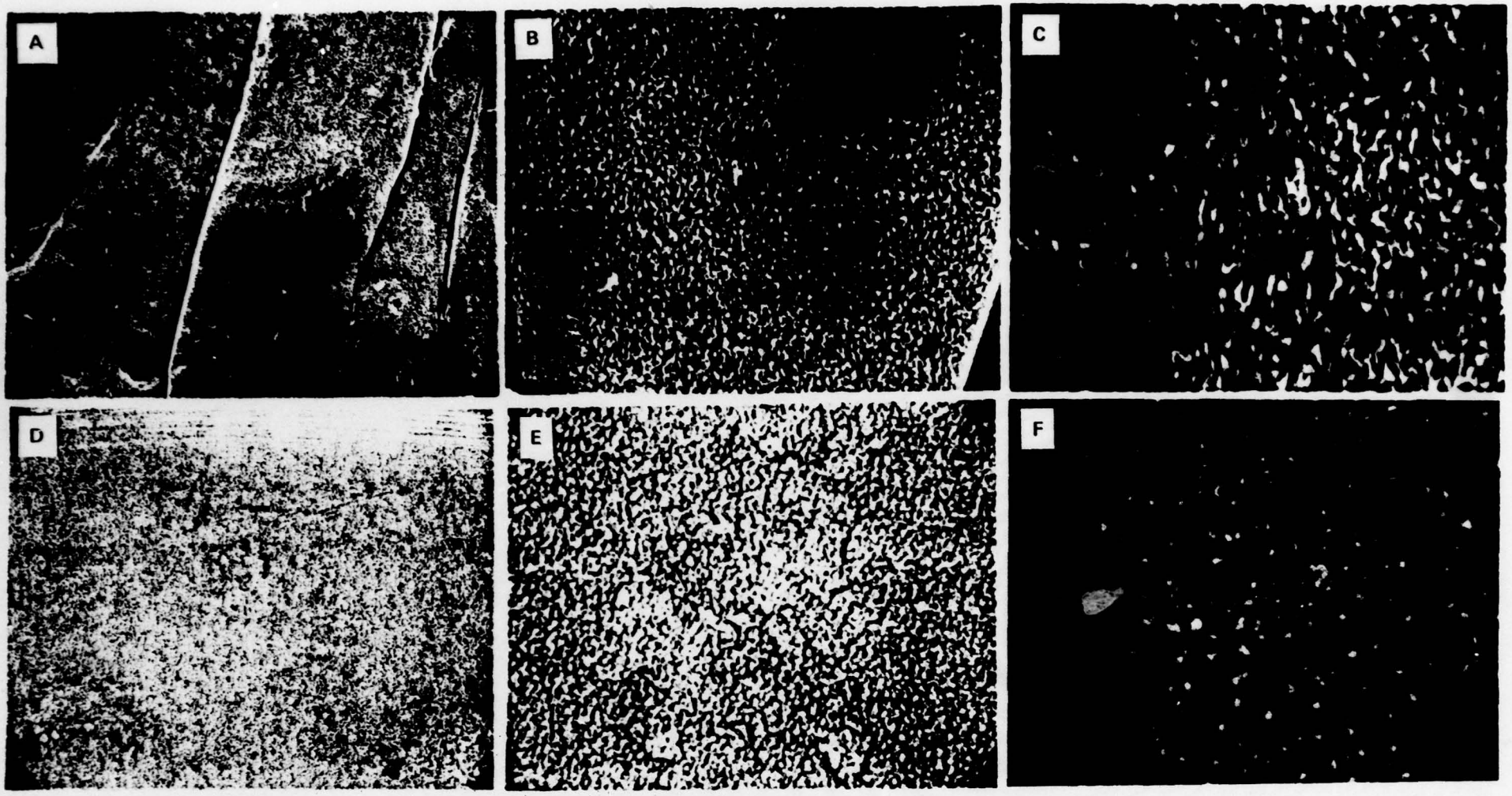


Figure B2. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = .25 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

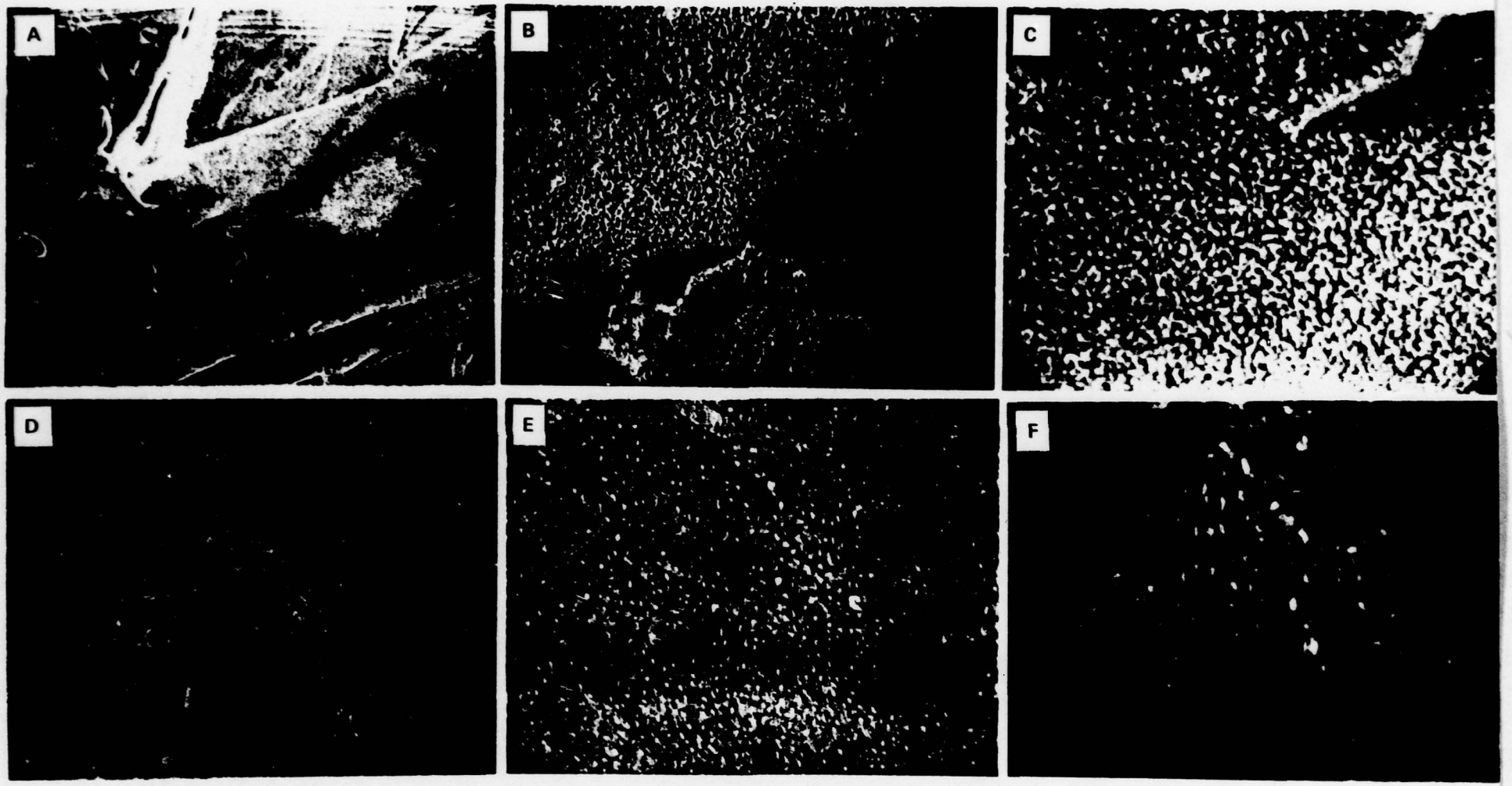


Figure B3. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = .5 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

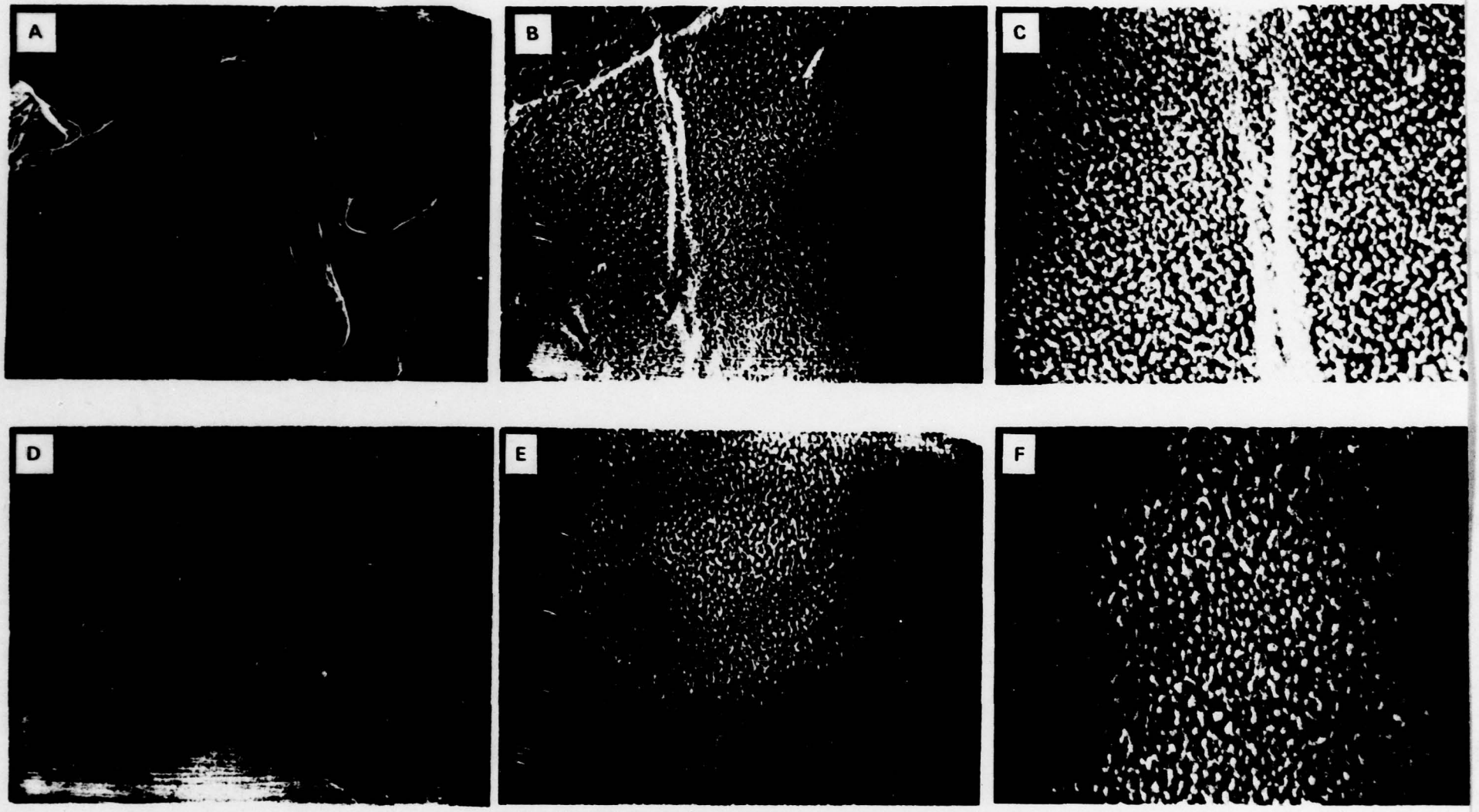


Figure B4. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 1 Day; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

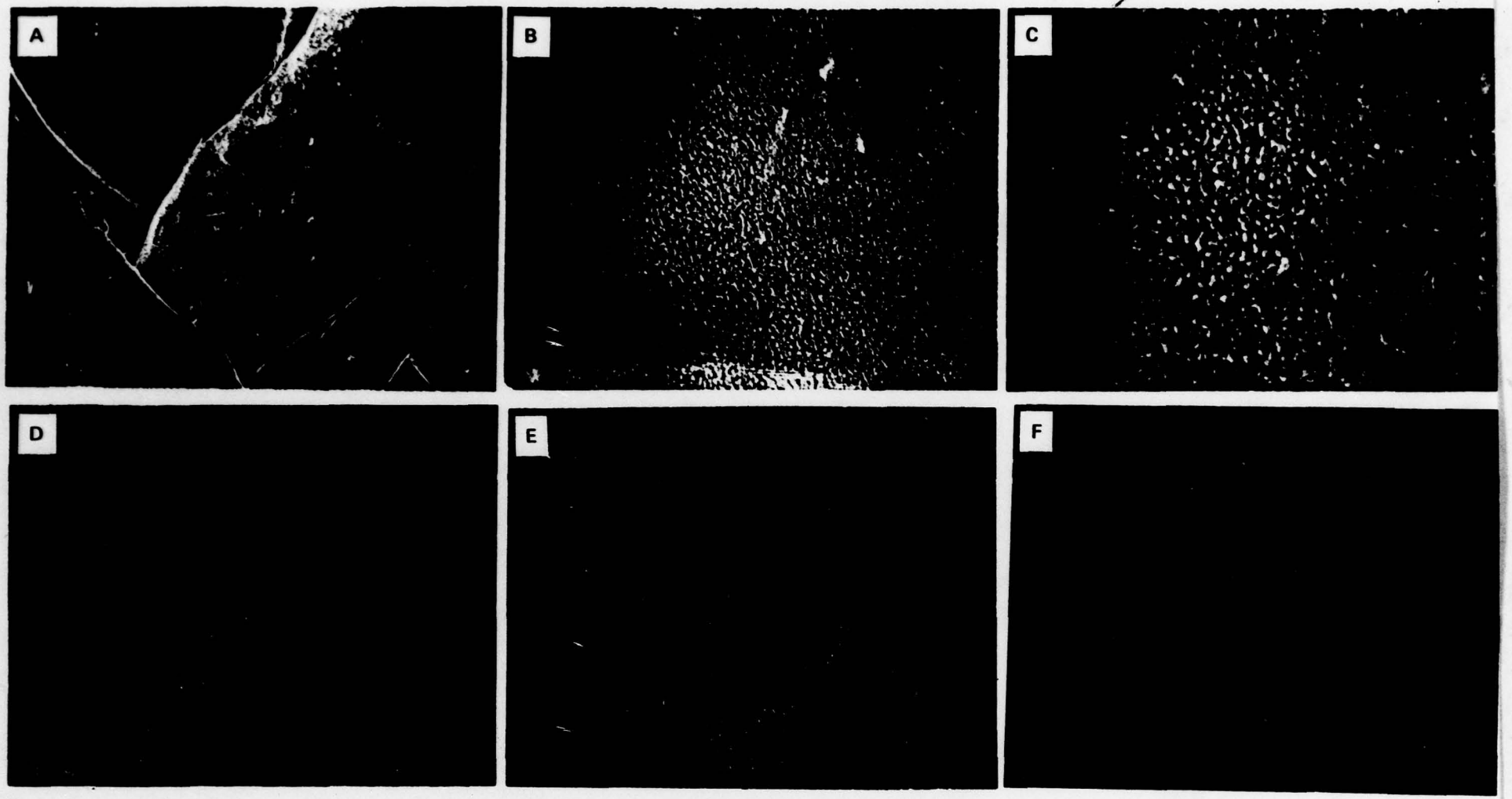


Figure B5. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 2 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

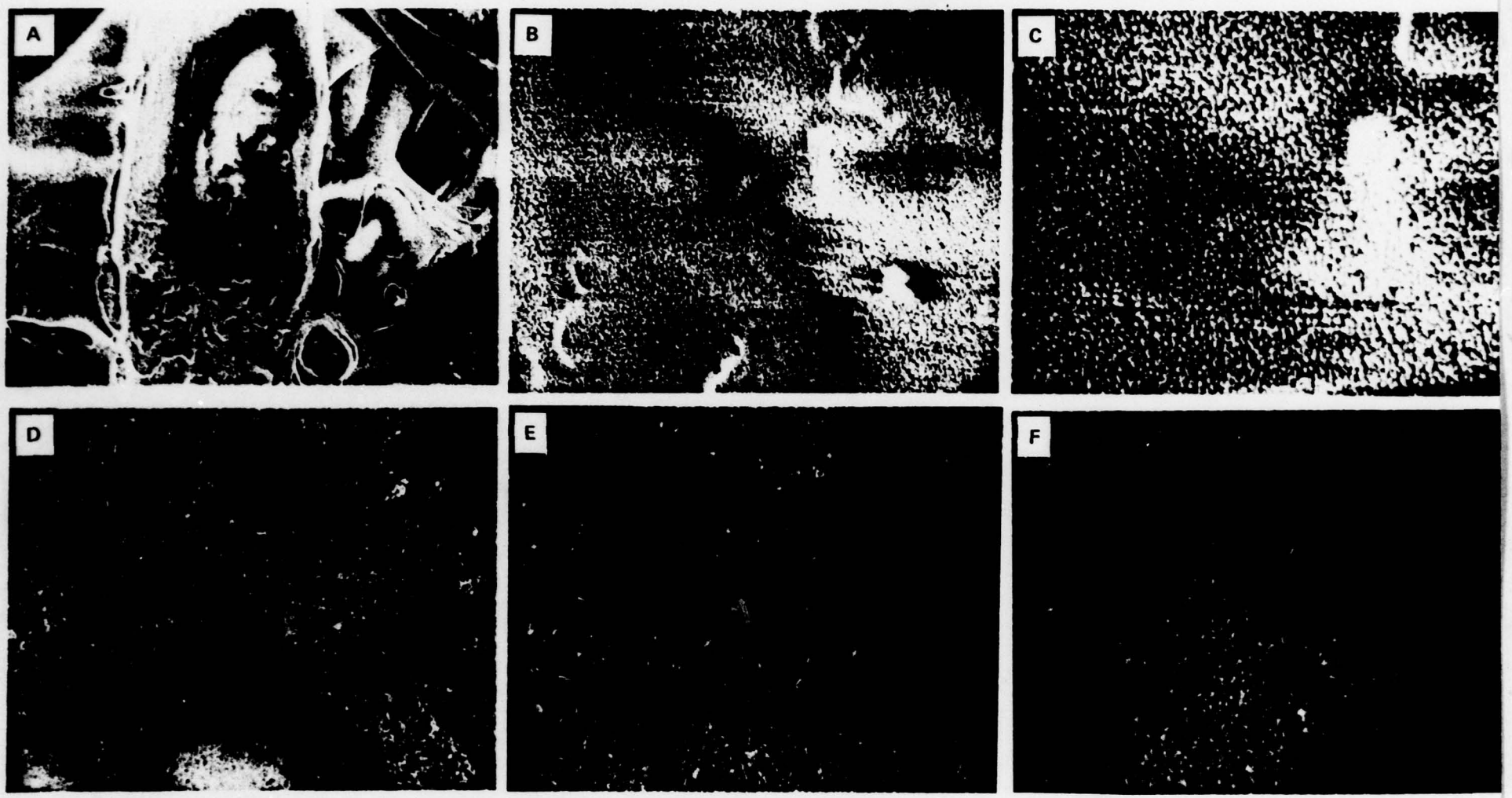
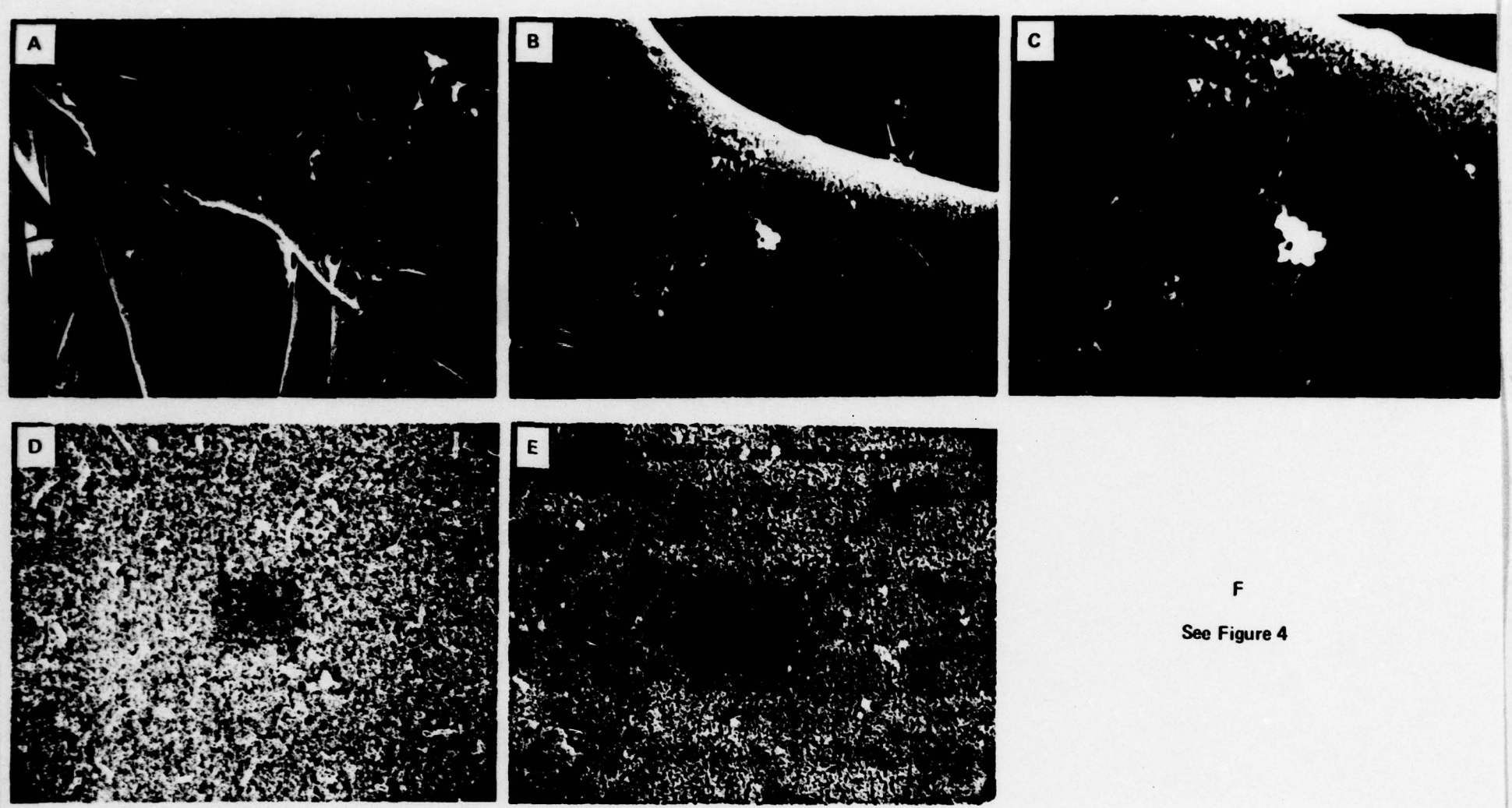


Figure B6. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 3 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



F

See Figure 4

Figure B7. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 5 Days; *Iodine Concentration* = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

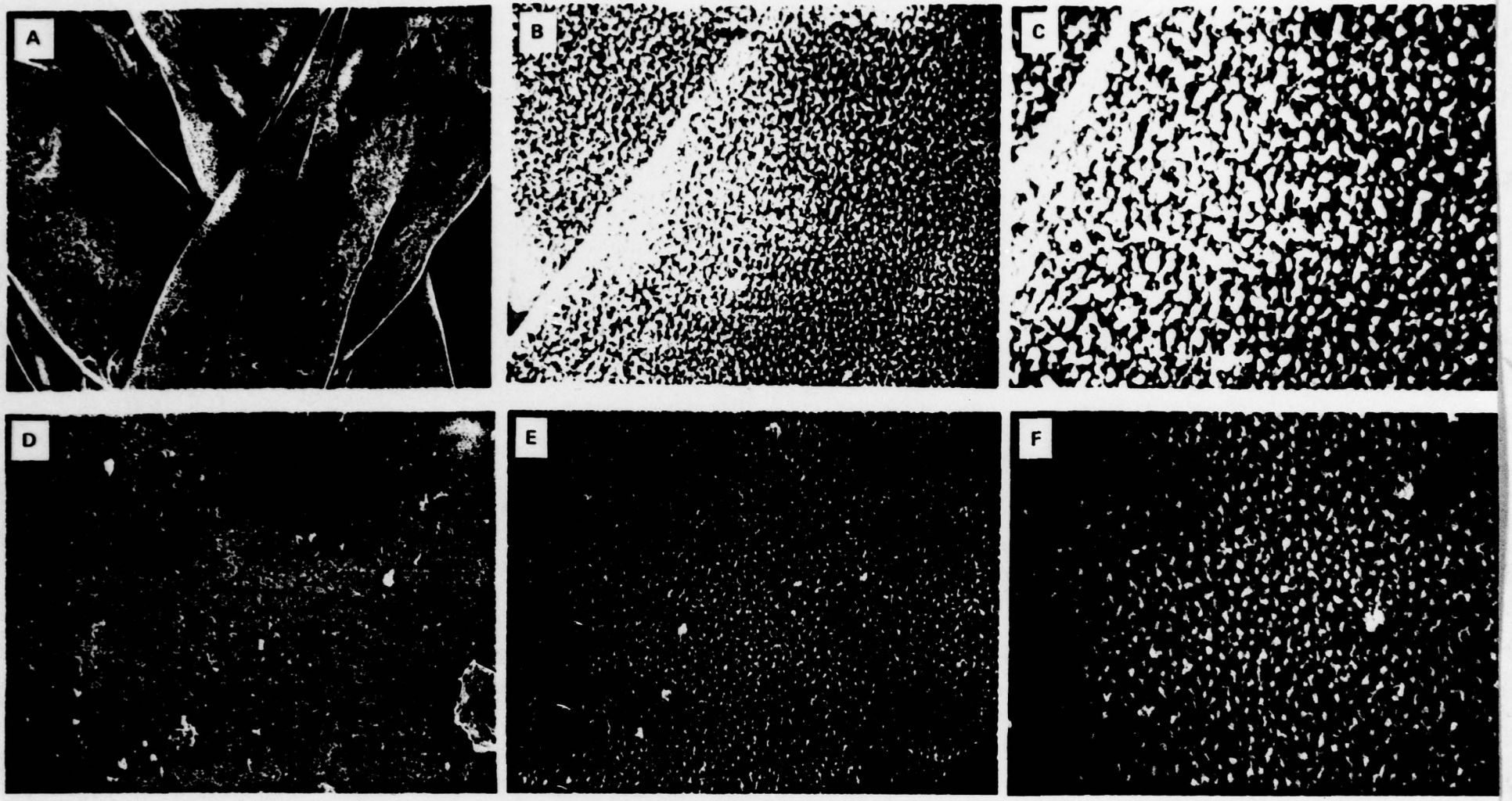


Figure B8. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 10 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

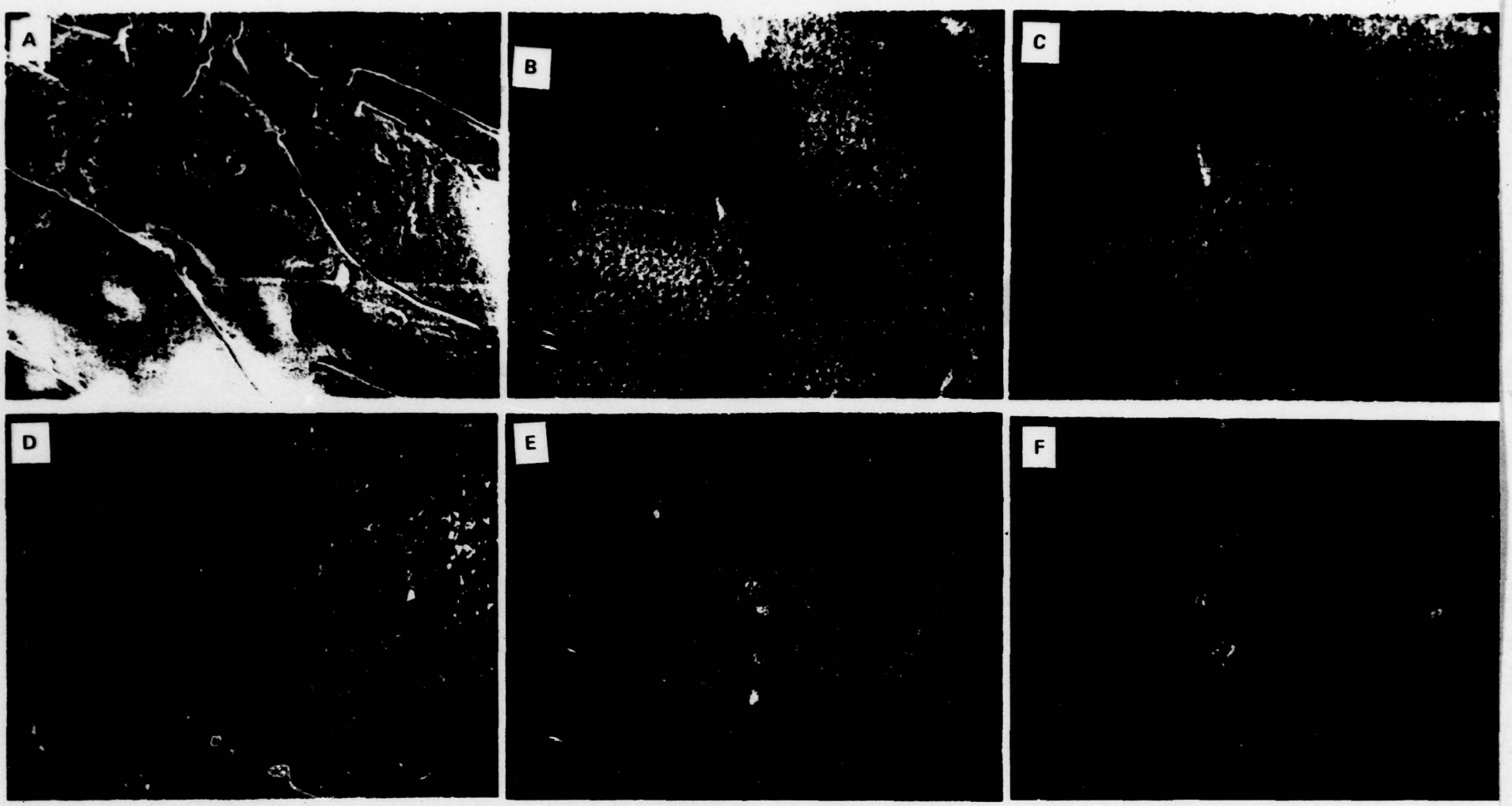
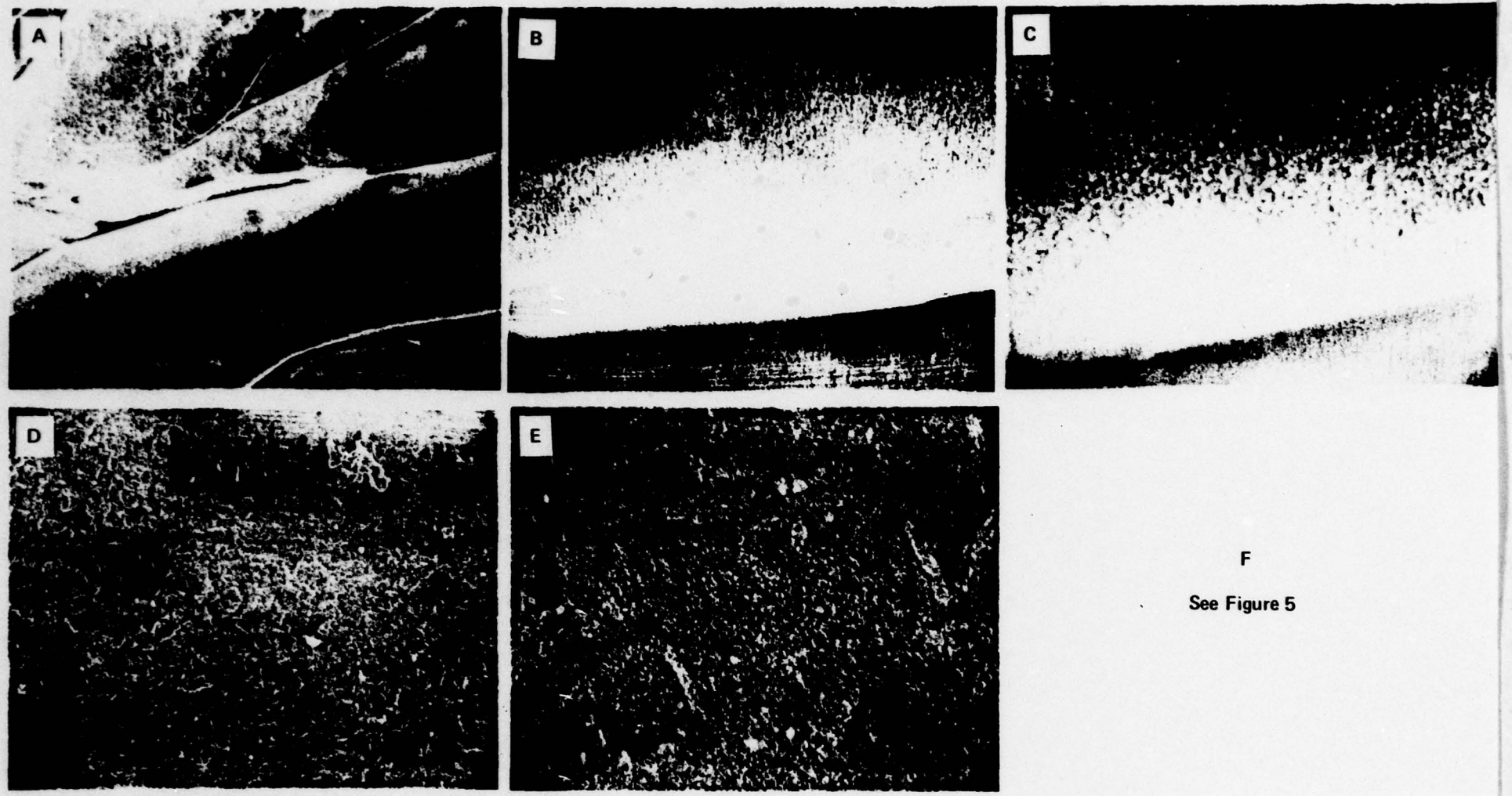


Figure B9. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 30 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



F

See Figure 5

Figure B10. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 45 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

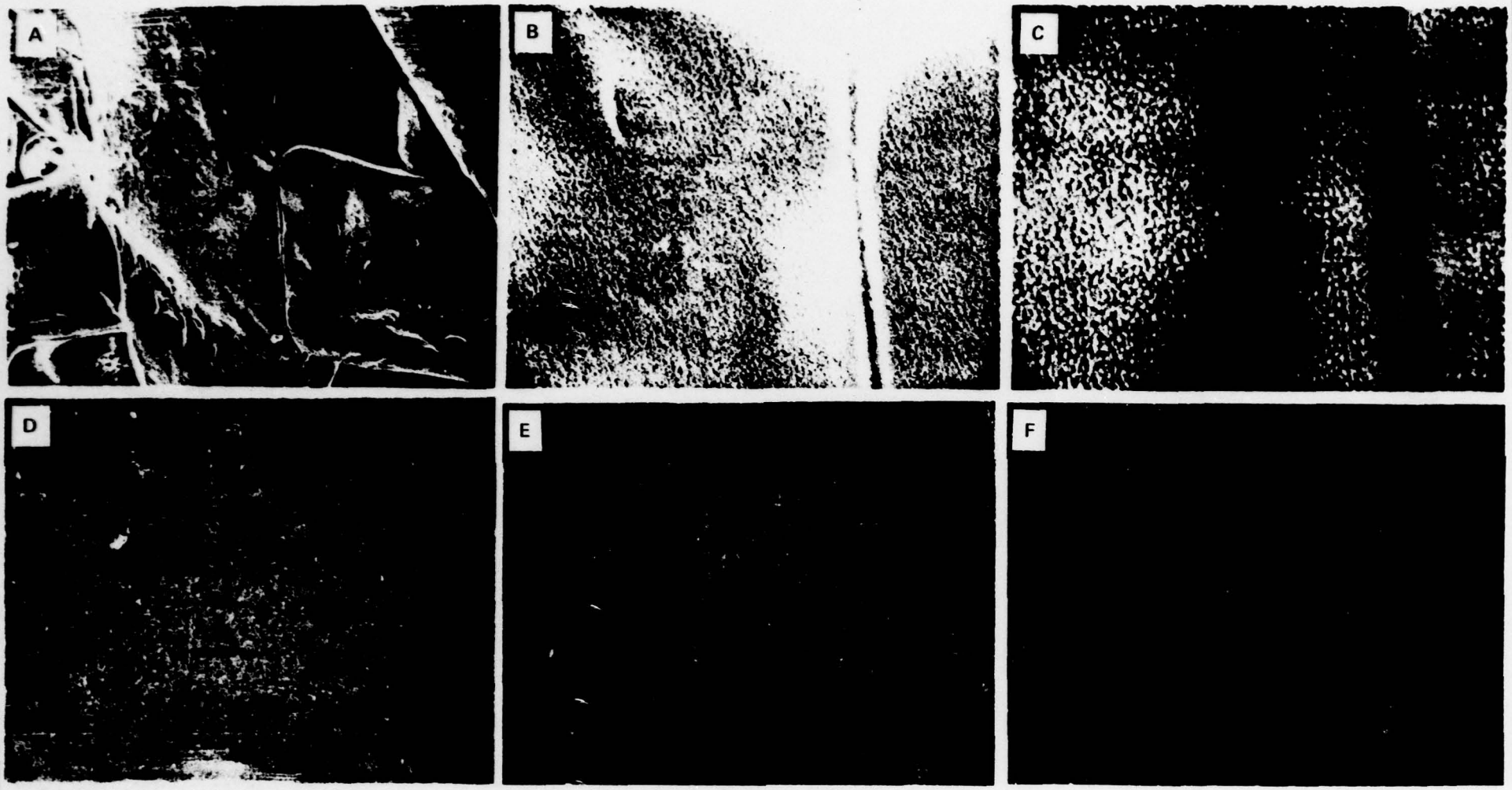
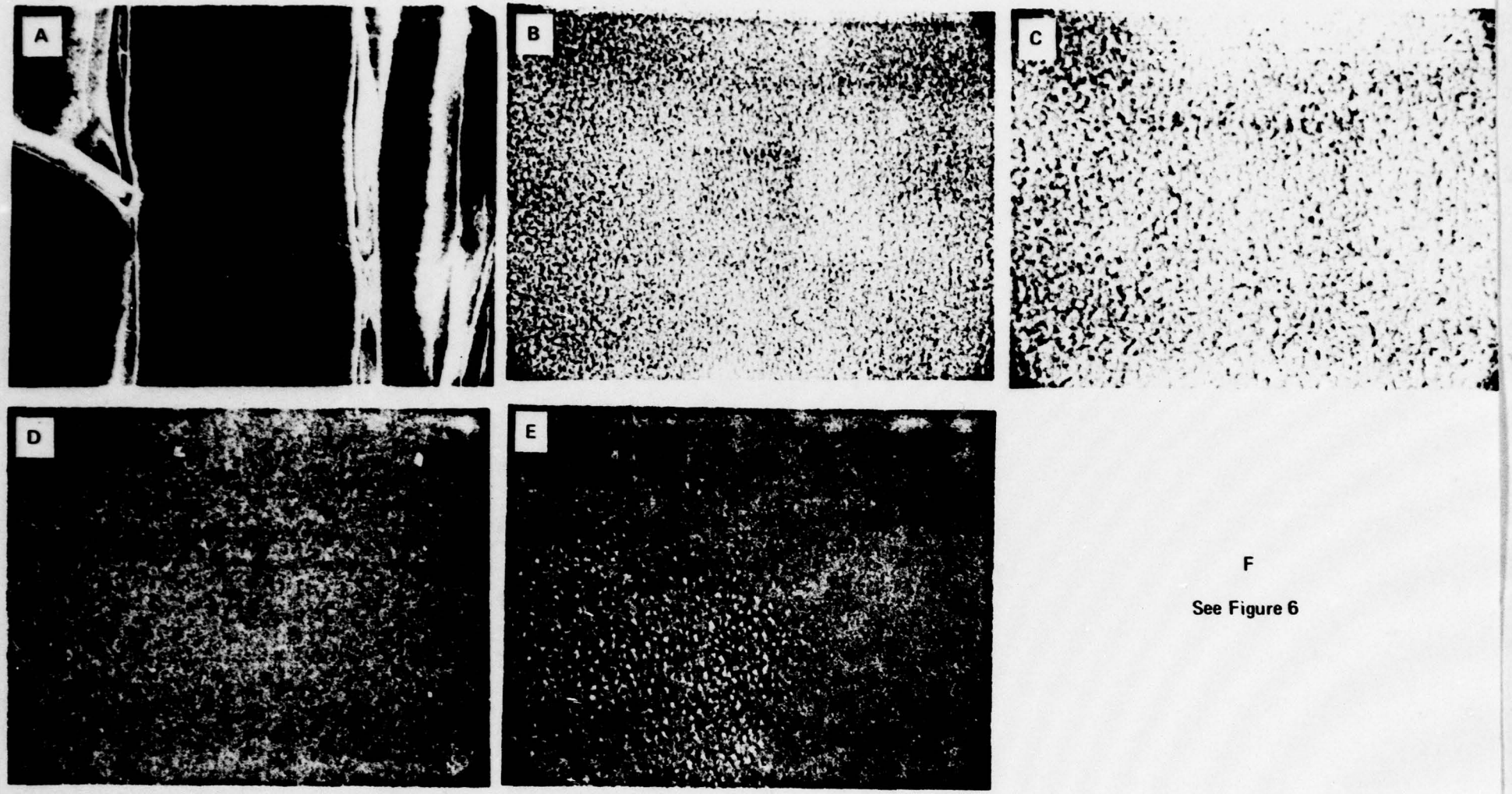


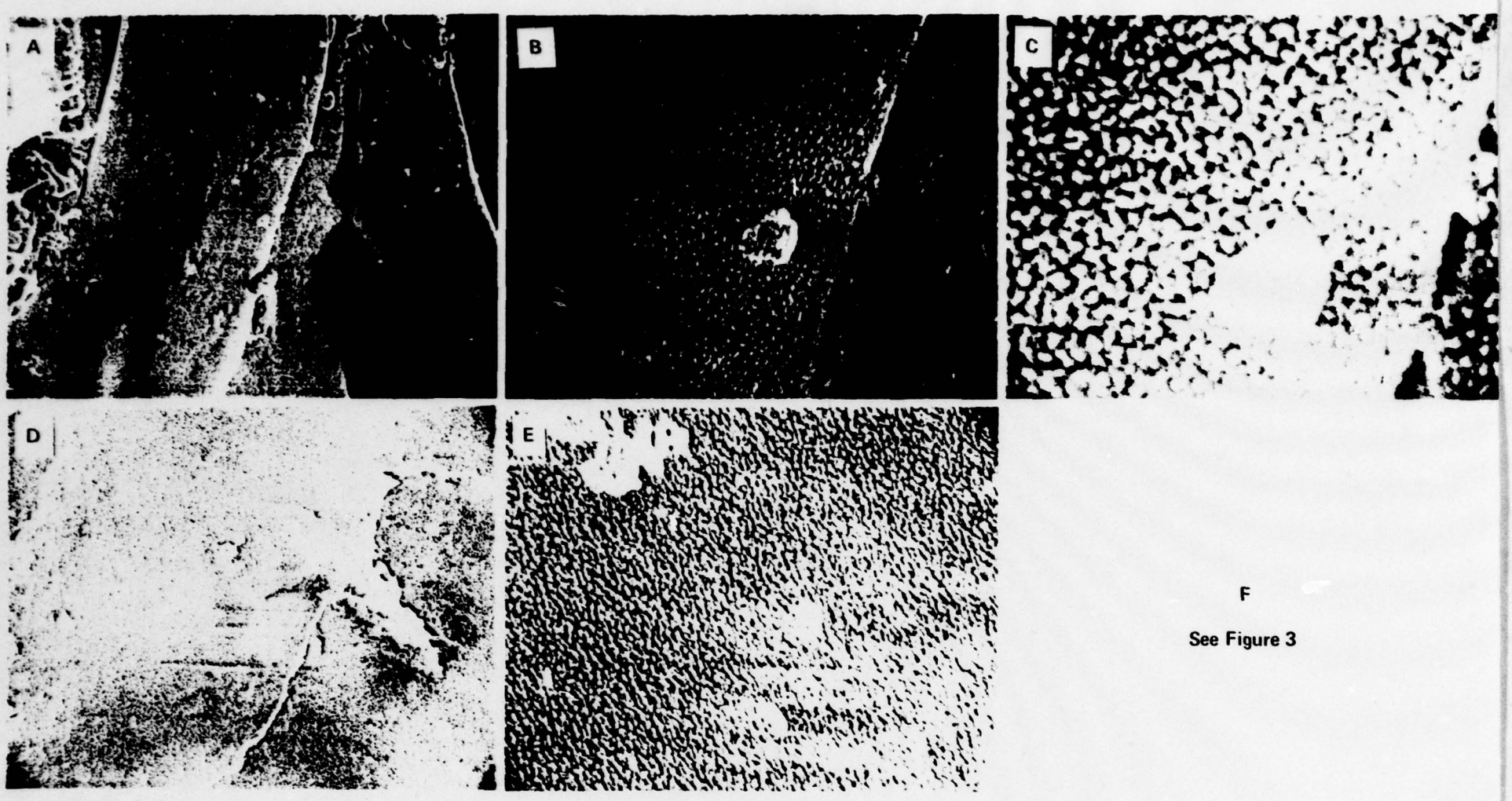
Figure B11. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 60 Days, Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



F

See Figure 6

Figure B12. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 83 Days; Iodine Concentration = 0 ppm (Buffer Alone)
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



F

See Figure 3

Figure B13. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 0 Days; Iodine Concentration = 1 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

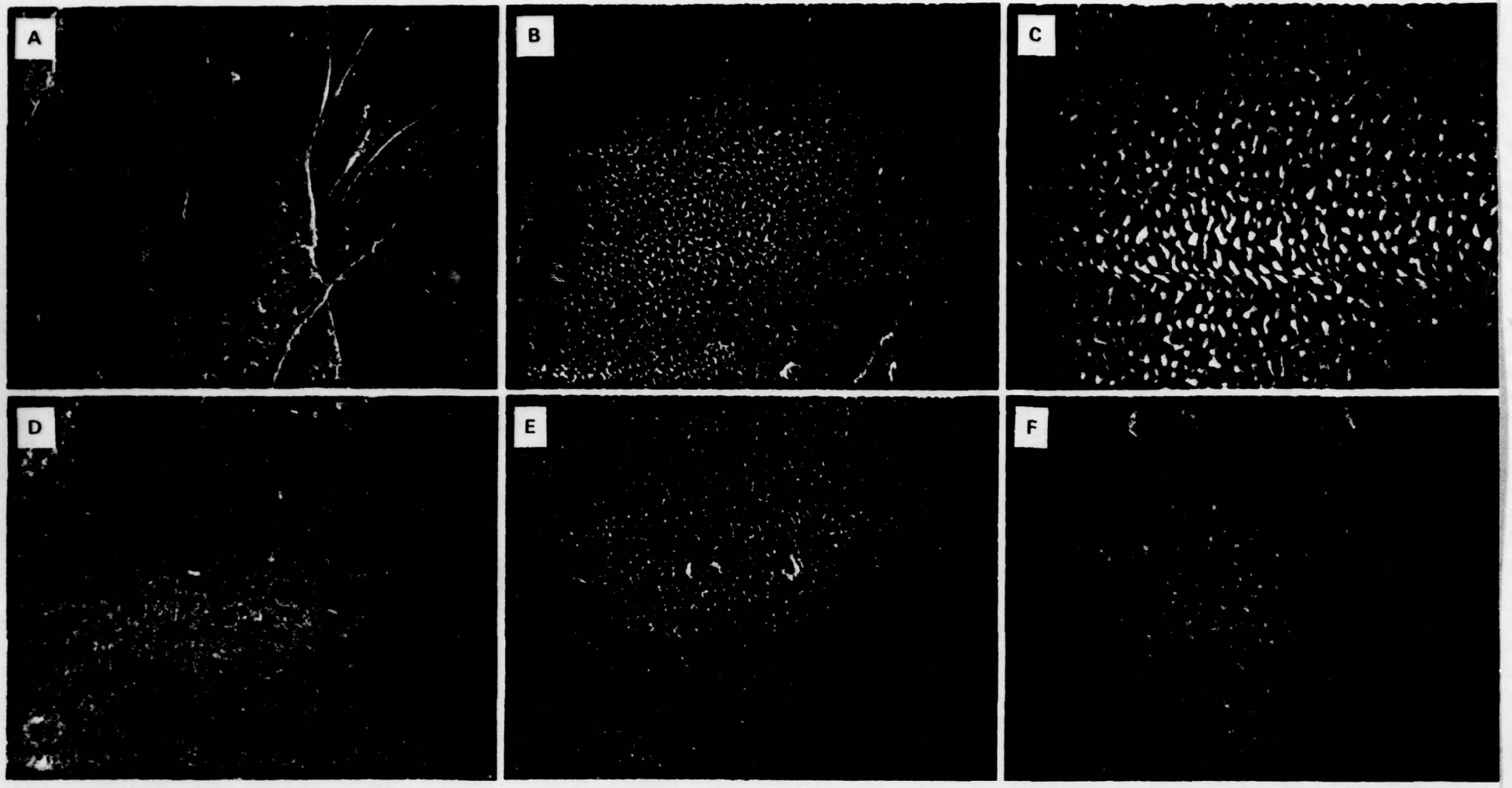
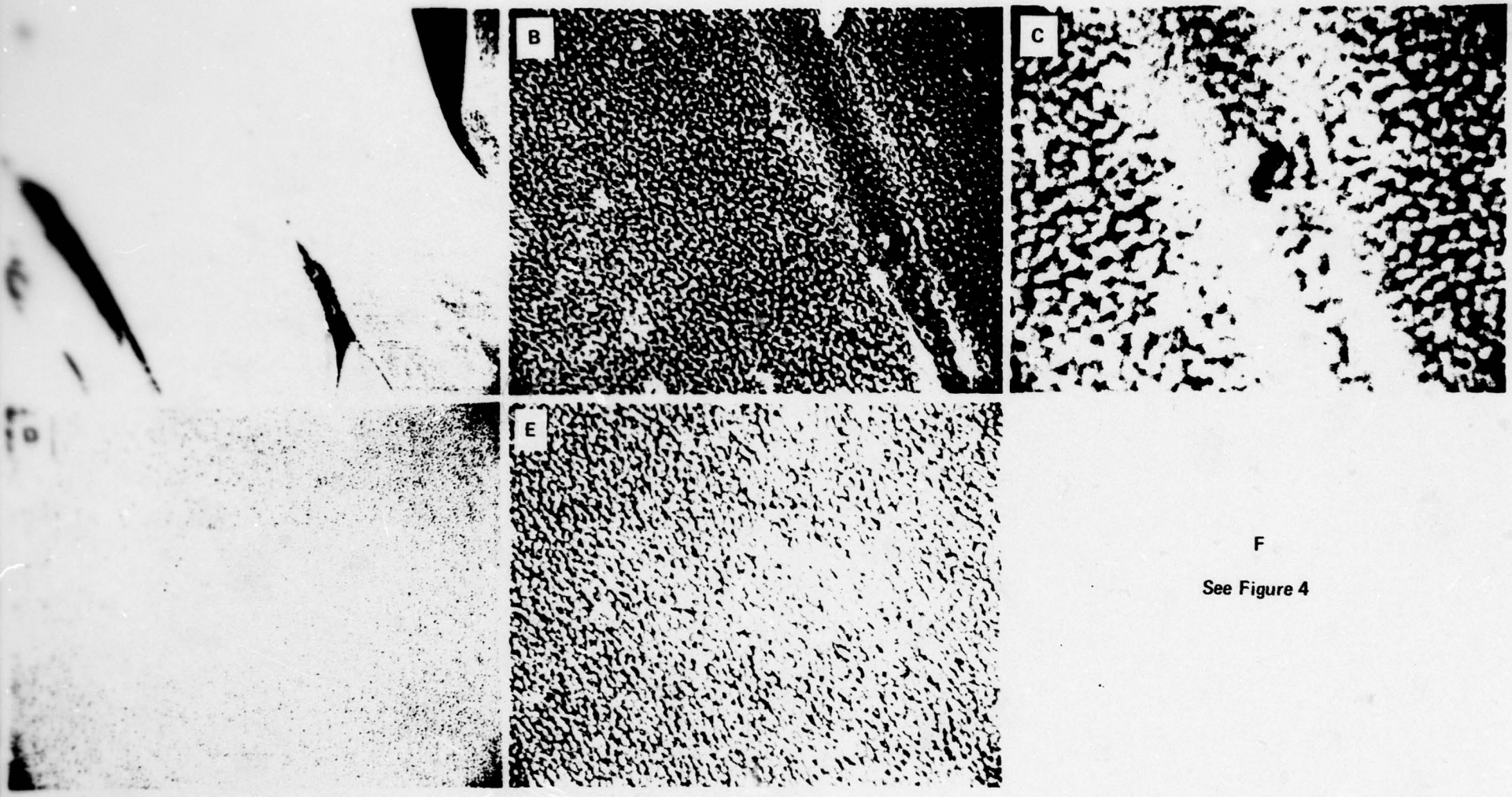


Figure B14. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 1 Day; Iodine Concentration = 1 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



See Figure 4

Figure B15. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 5 Days; Iodine Concentration = 1 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

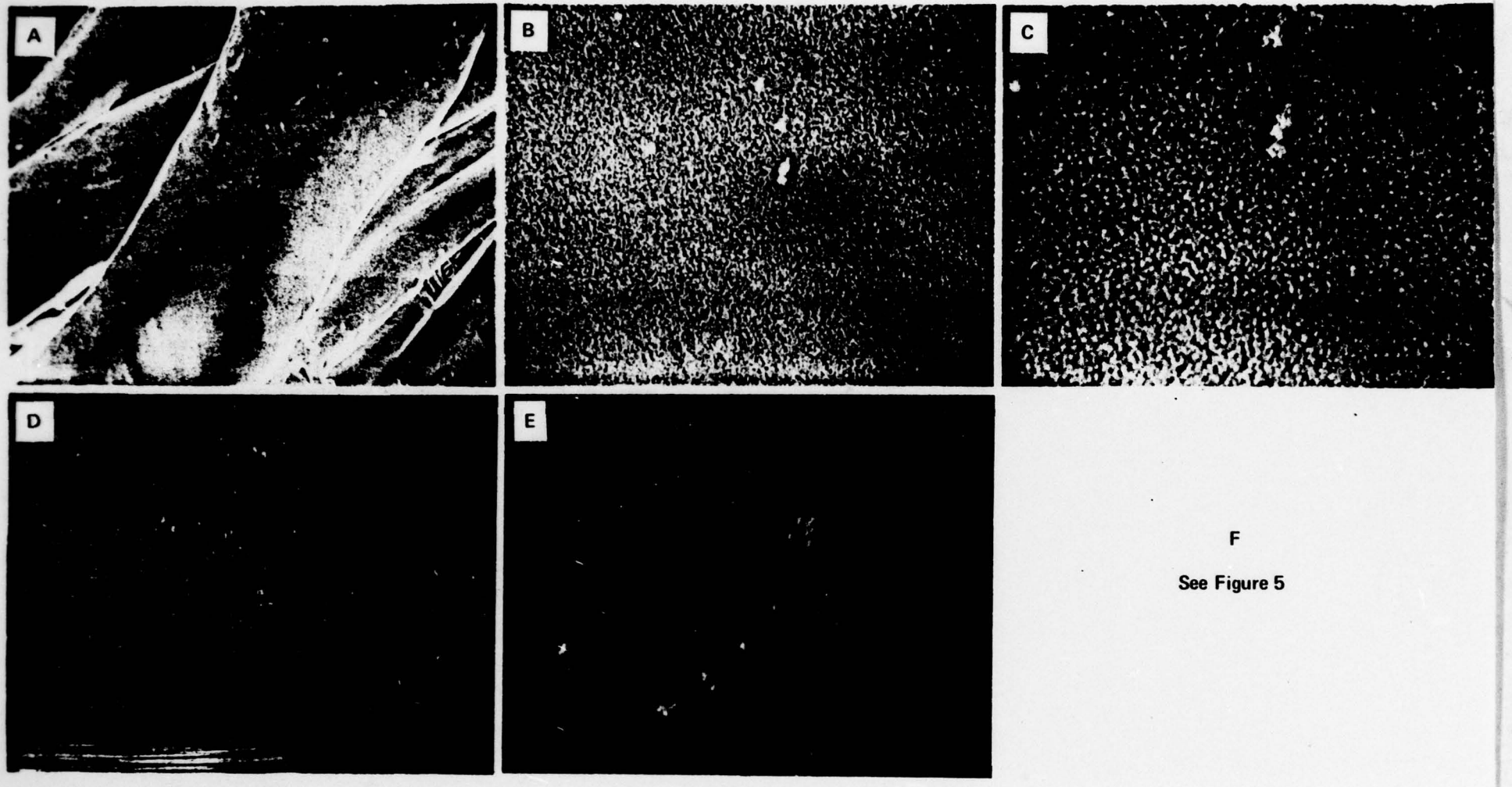
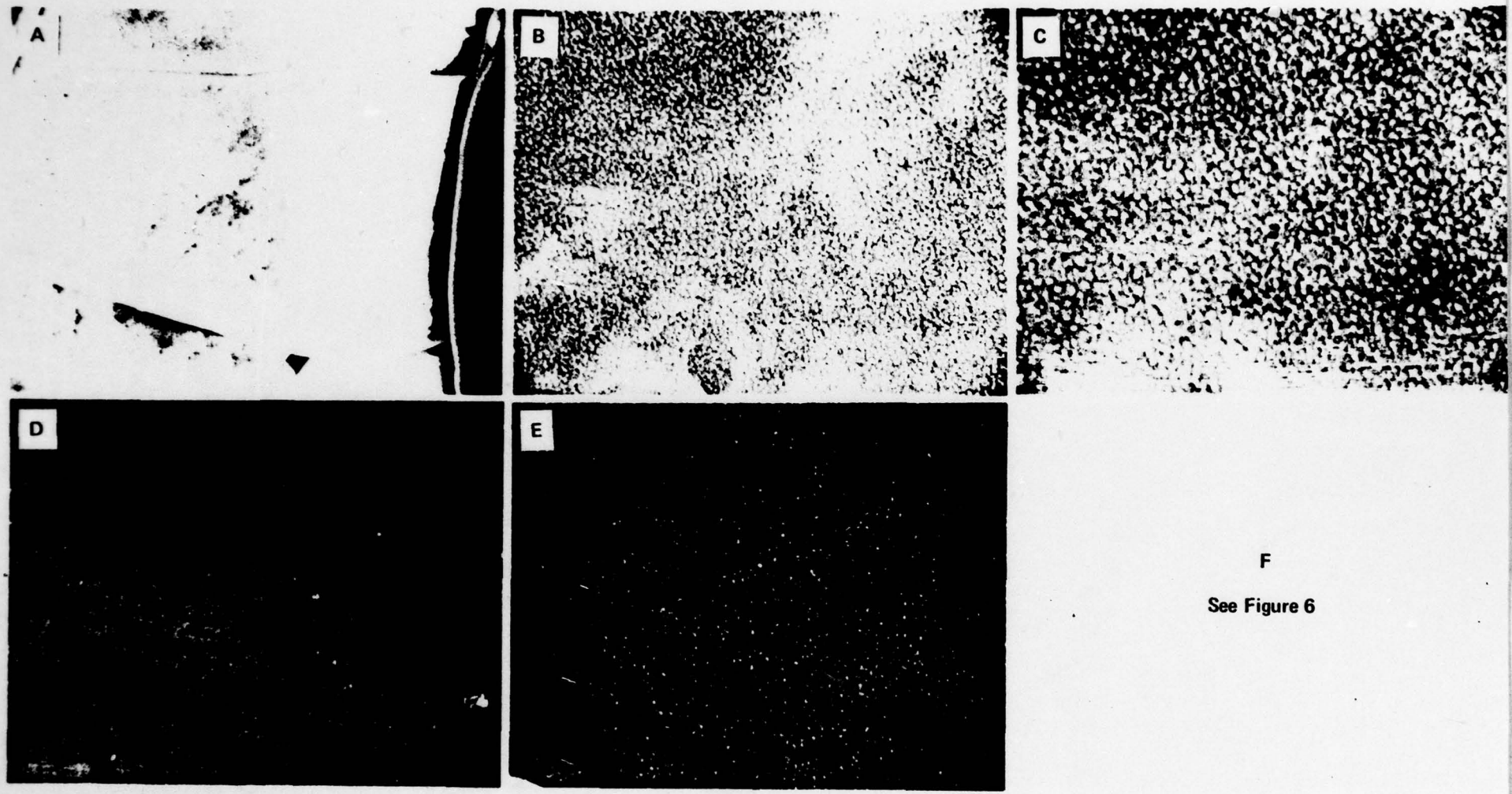


Figure B16. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 45 Days; Iodine Concentration = 1 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X



F

See Figure 6

Figure B17. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 83 Days; Iodine Concentration = 1 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

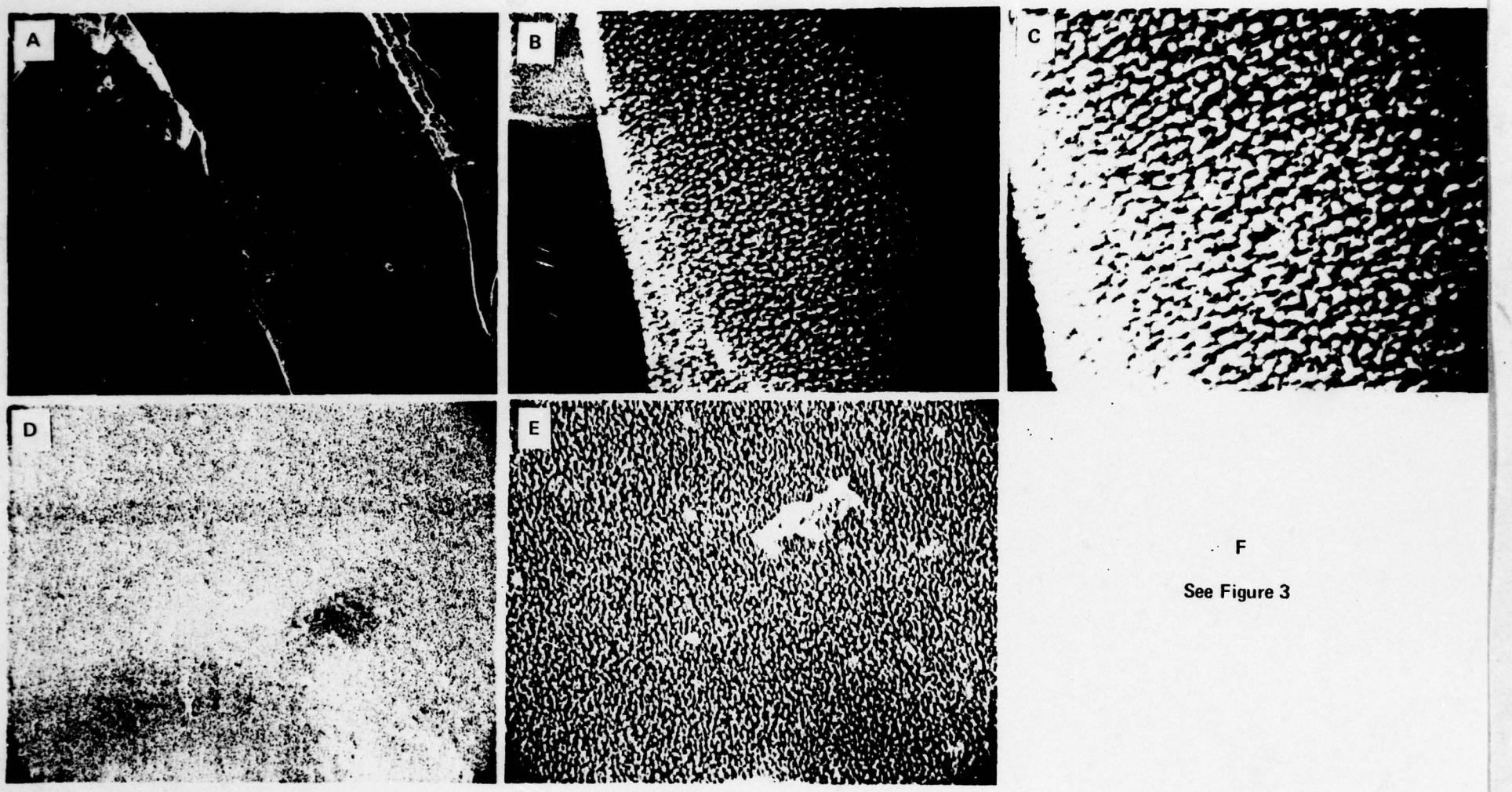


Figure B18. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 0 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

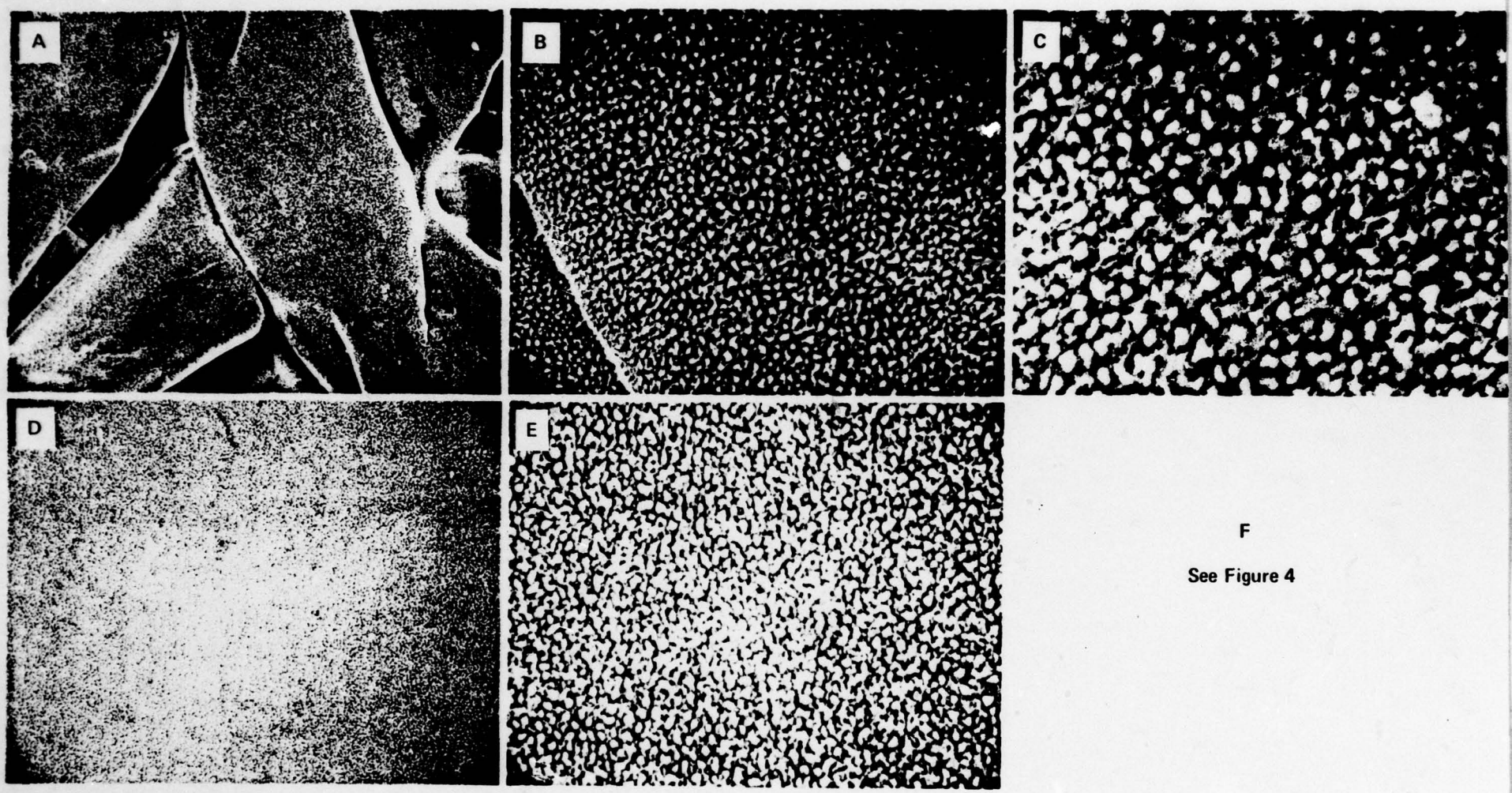


Figure B19. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 5 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

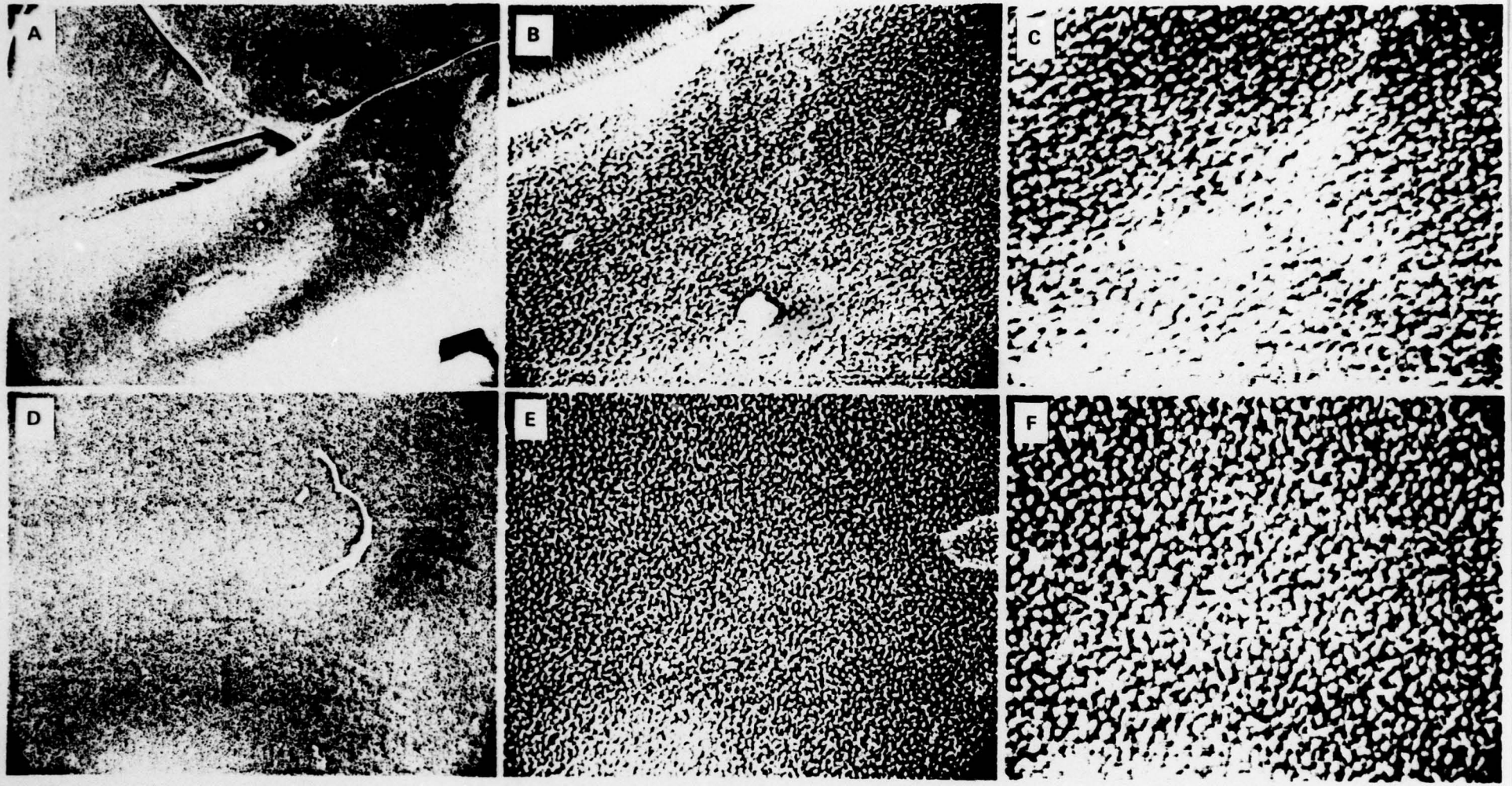


Figure B20. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 10 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

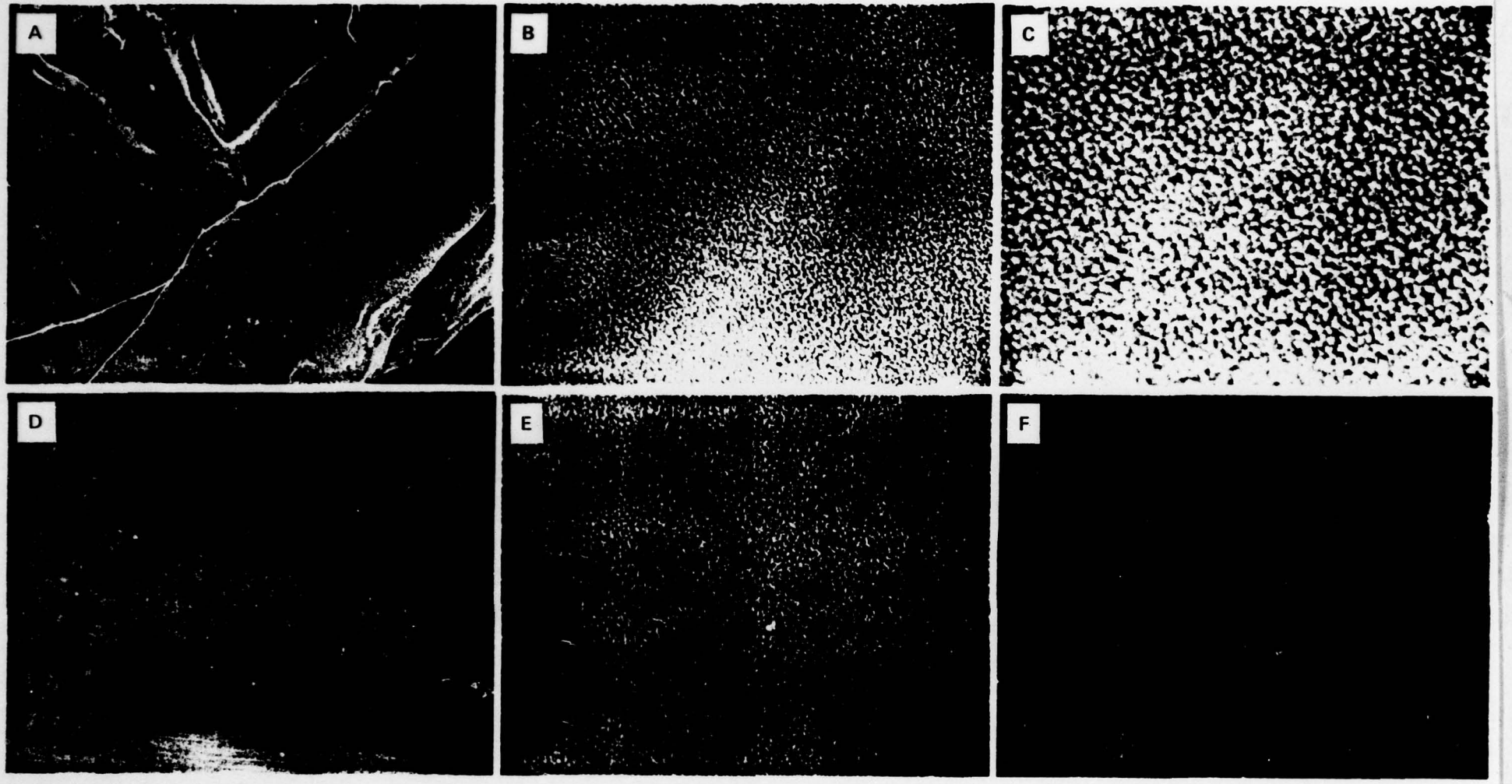


Figure B21. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 30 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

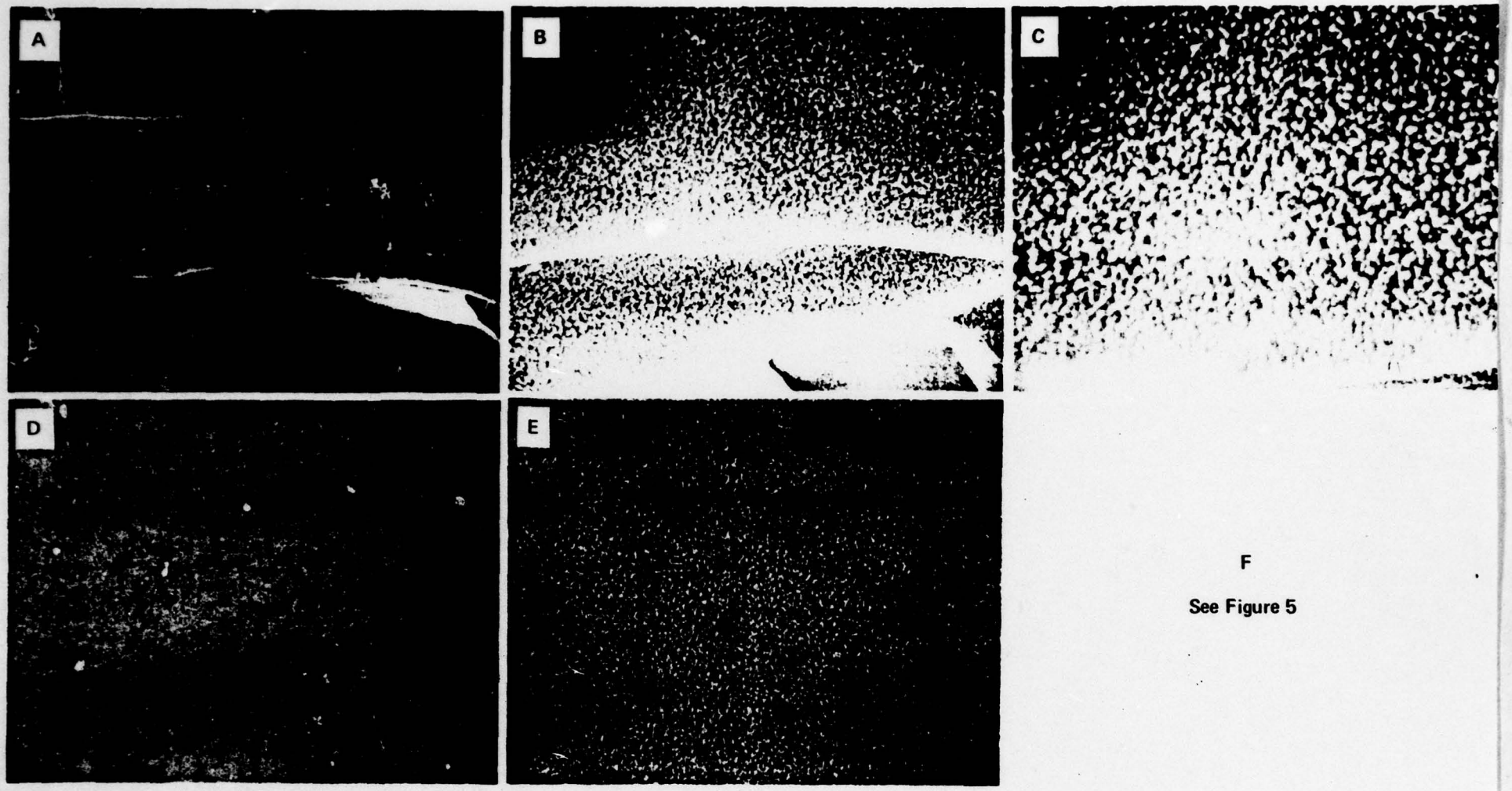


Figure B22. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 45 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

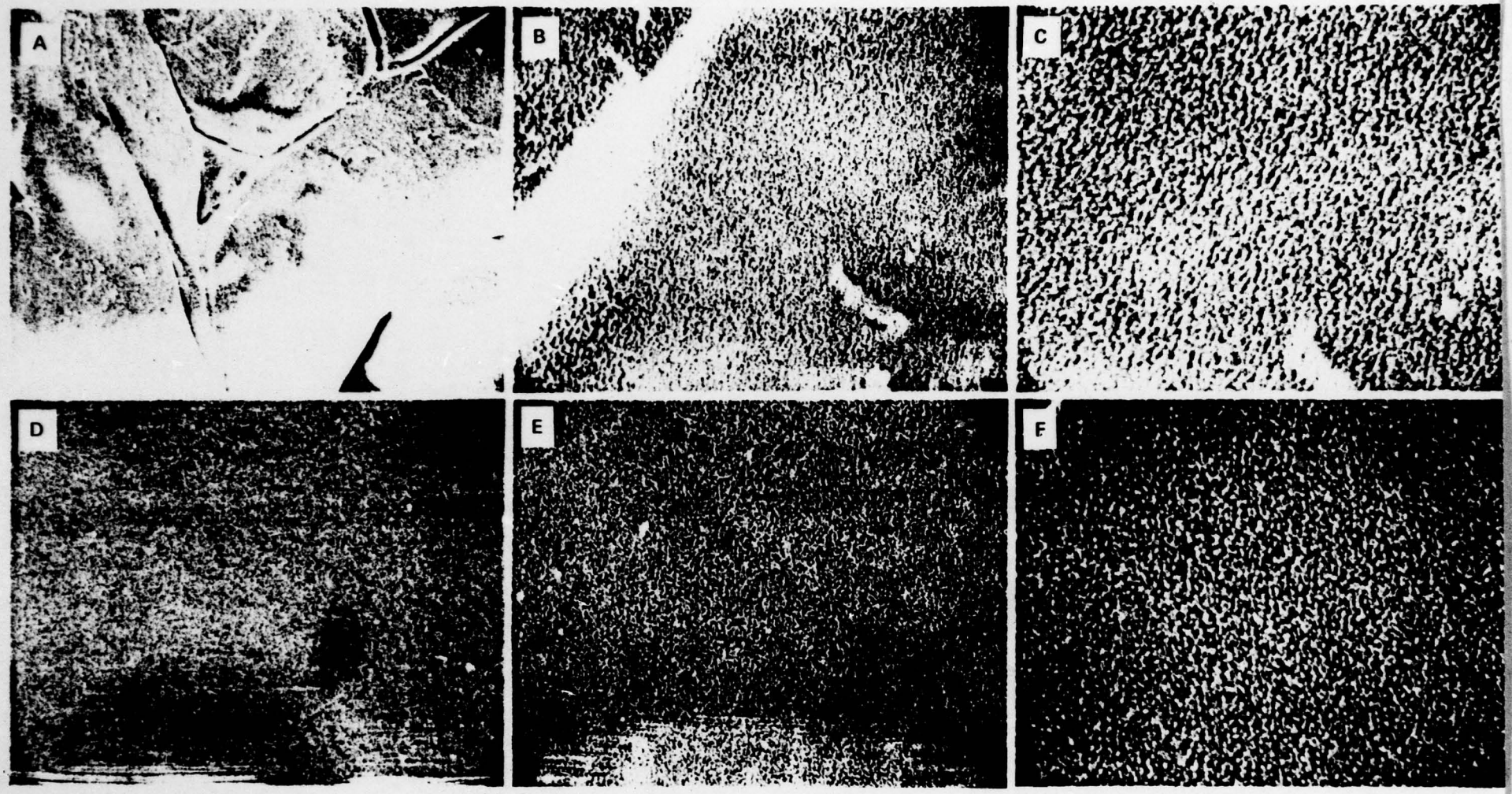


Figure B23. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 60 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

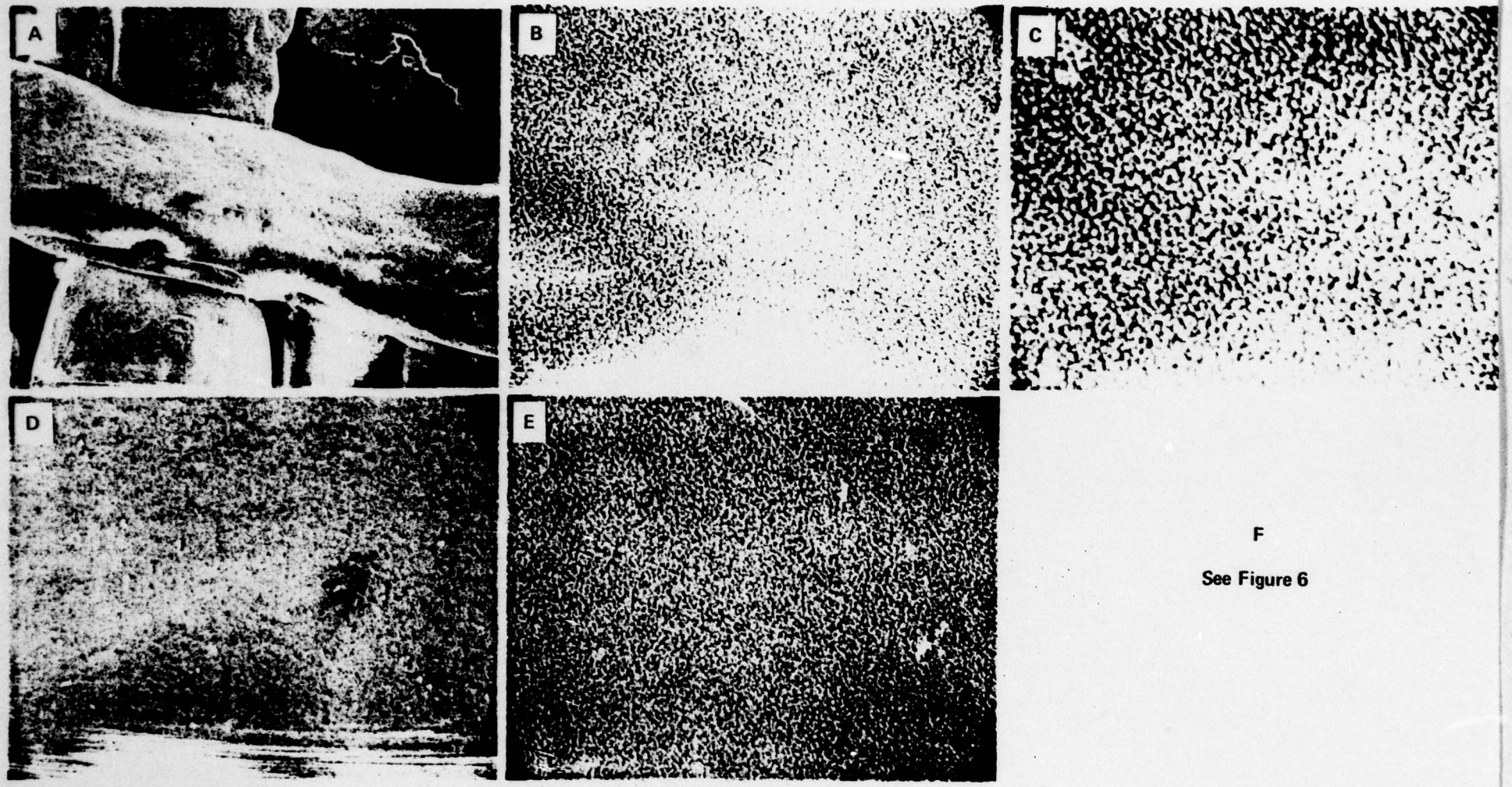


Figure B24. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 83 Days; Iodine Concentration = 5 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

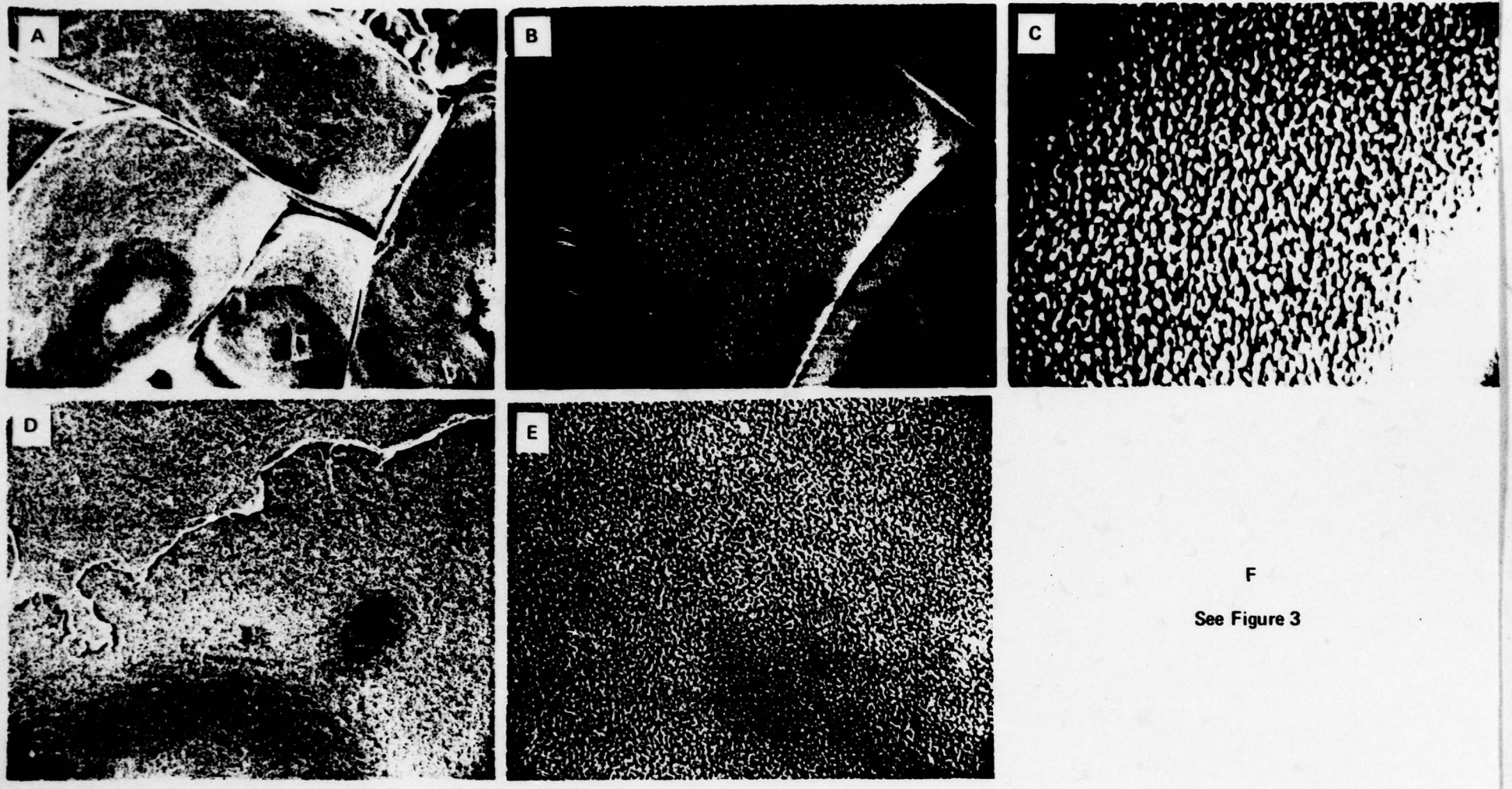


Figure B25. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 0 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

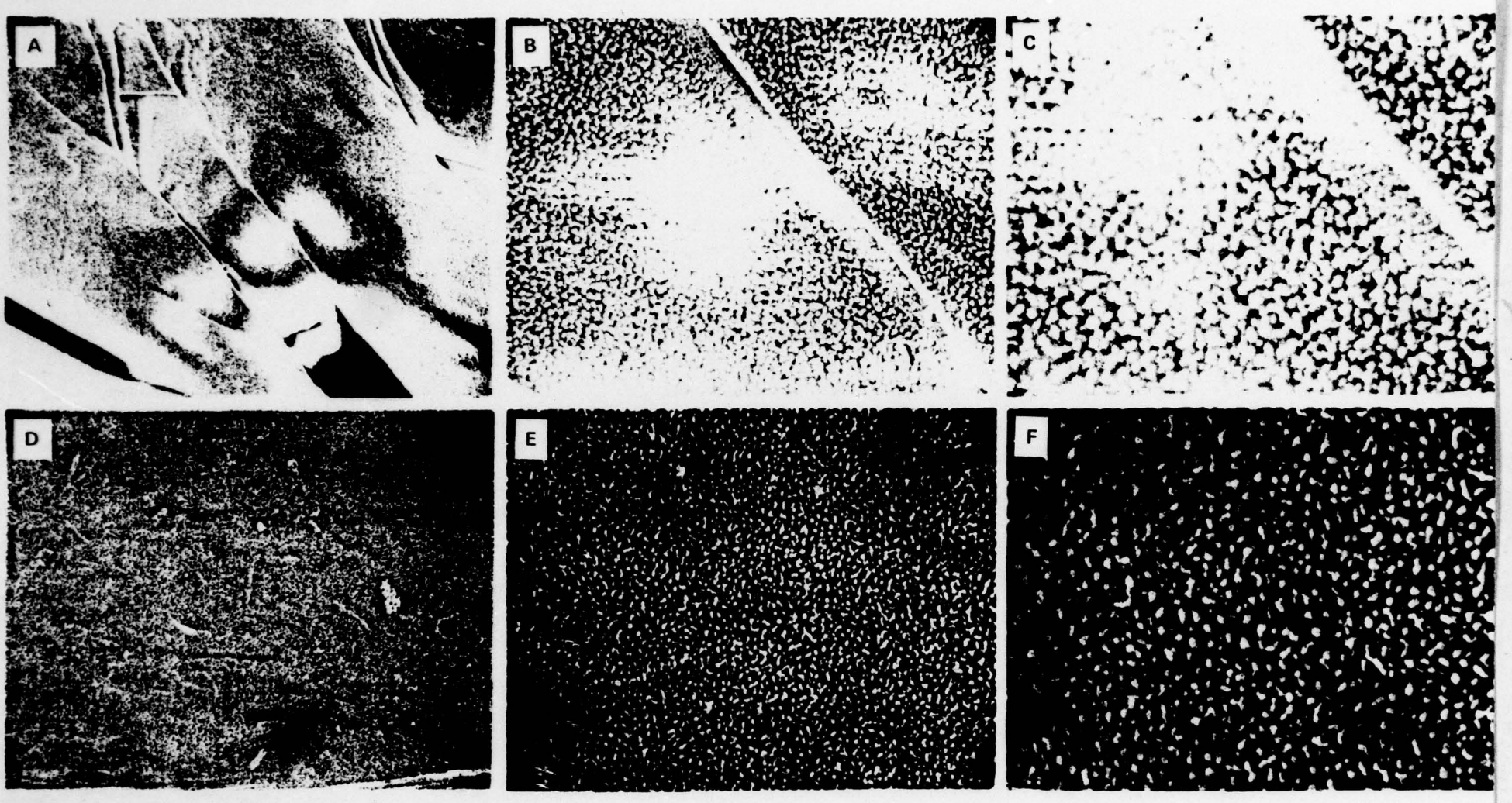


Figure B26. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = .25 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

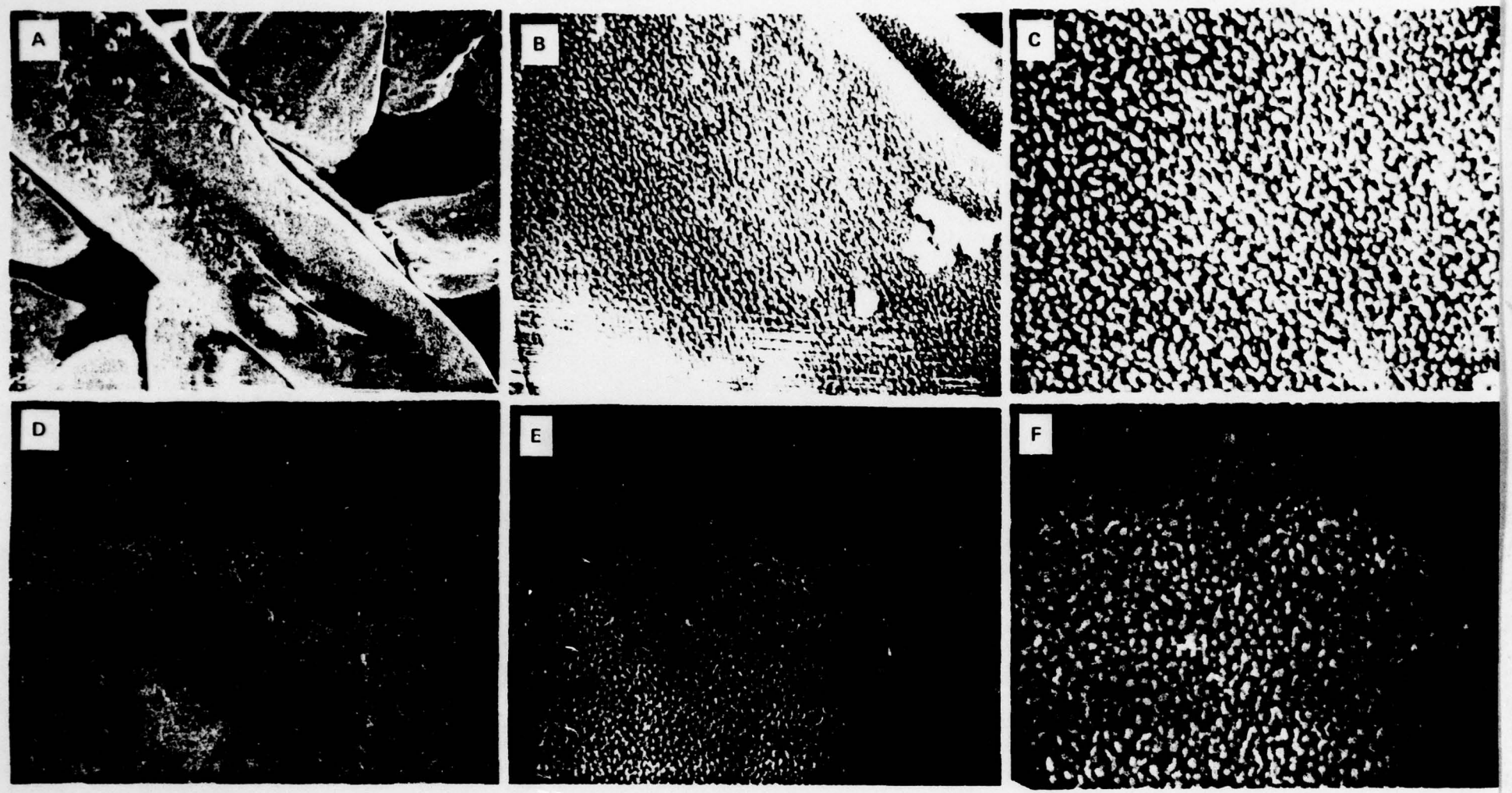


Figure B27. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = .5 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

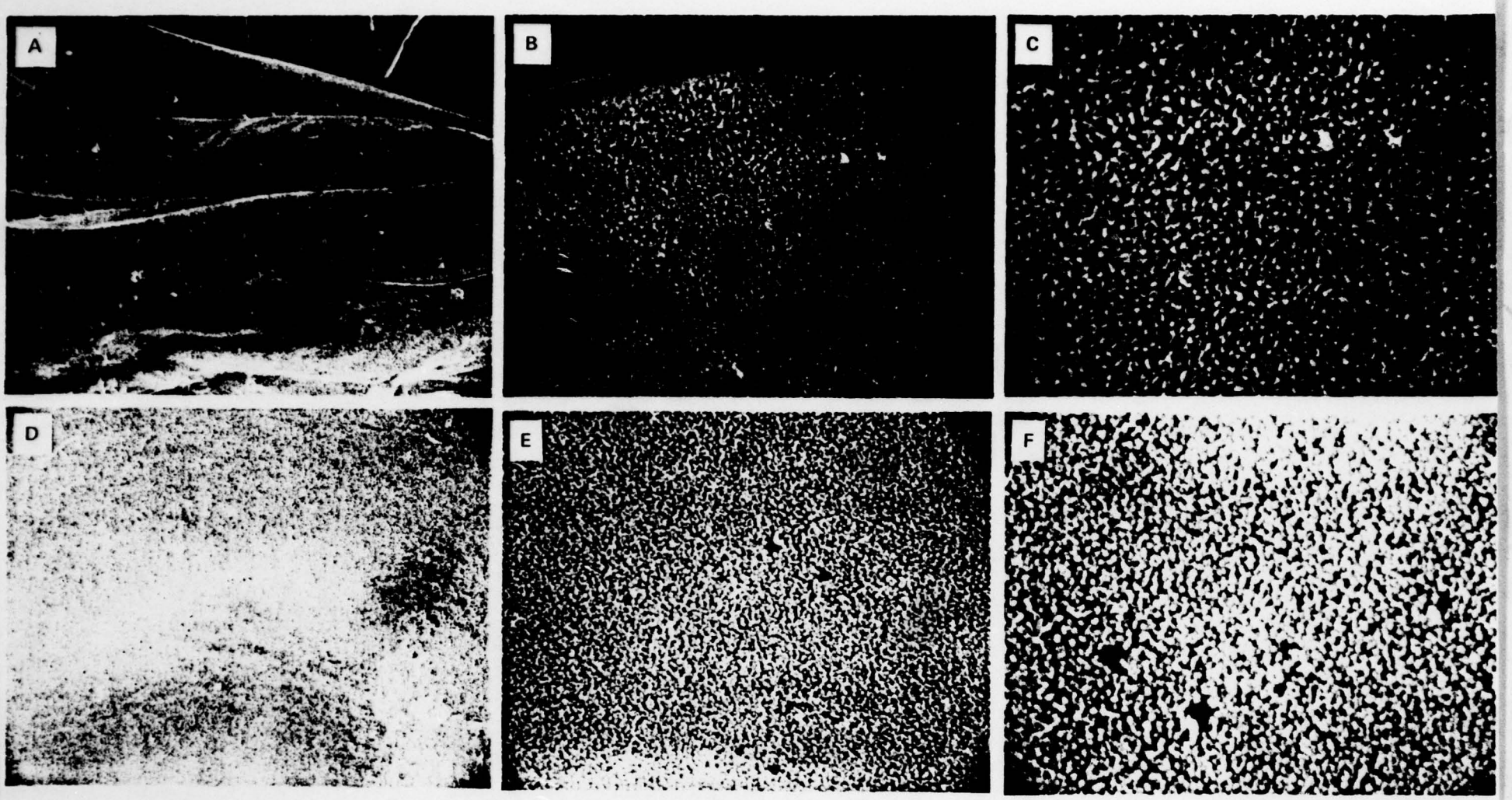


Figure B28. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 1 Day; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D -- 1,400X; E – 7,000X; F – 14,000X

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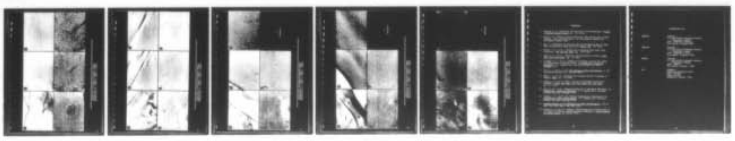

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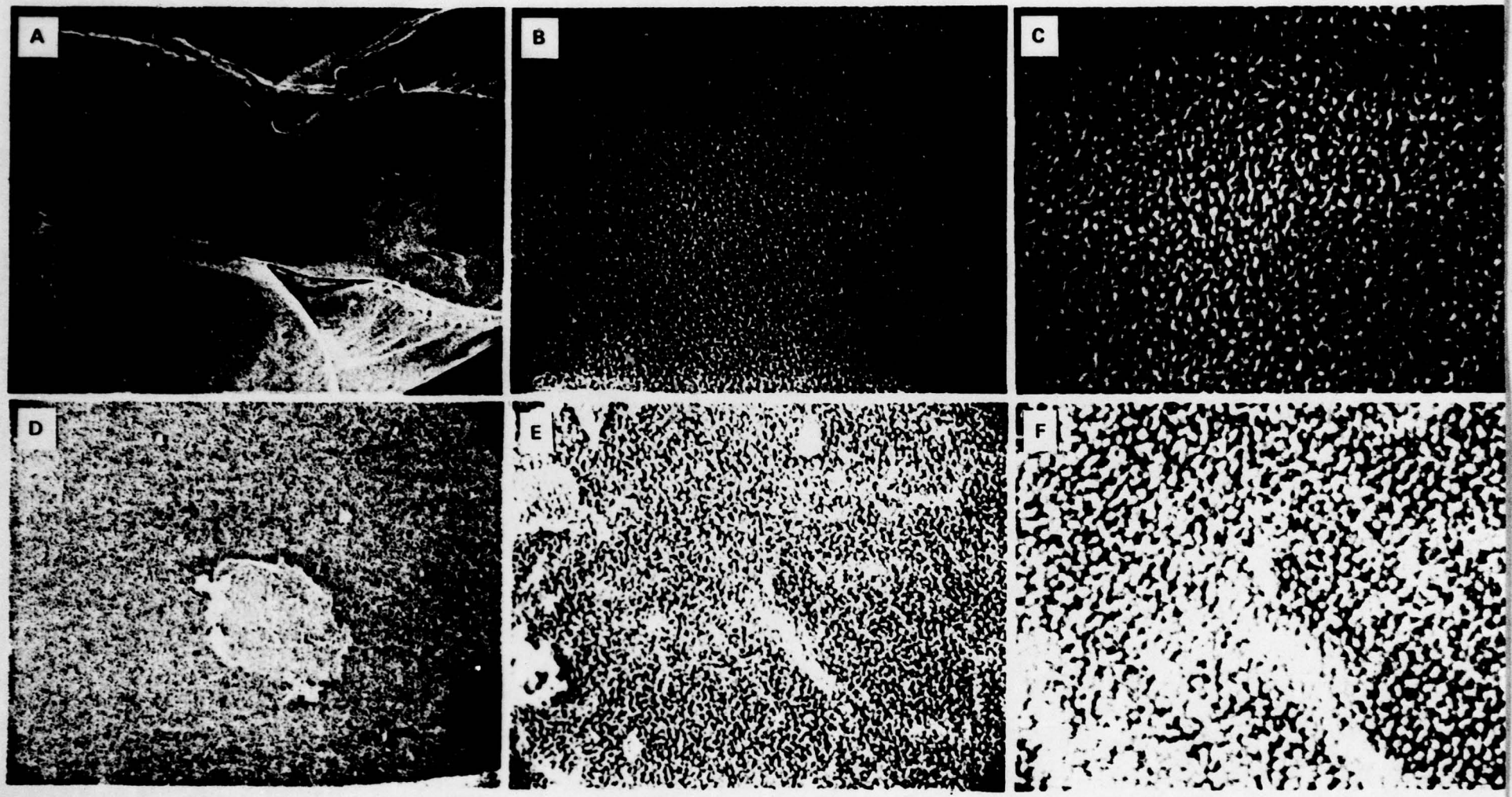

Figure B29. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 2 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

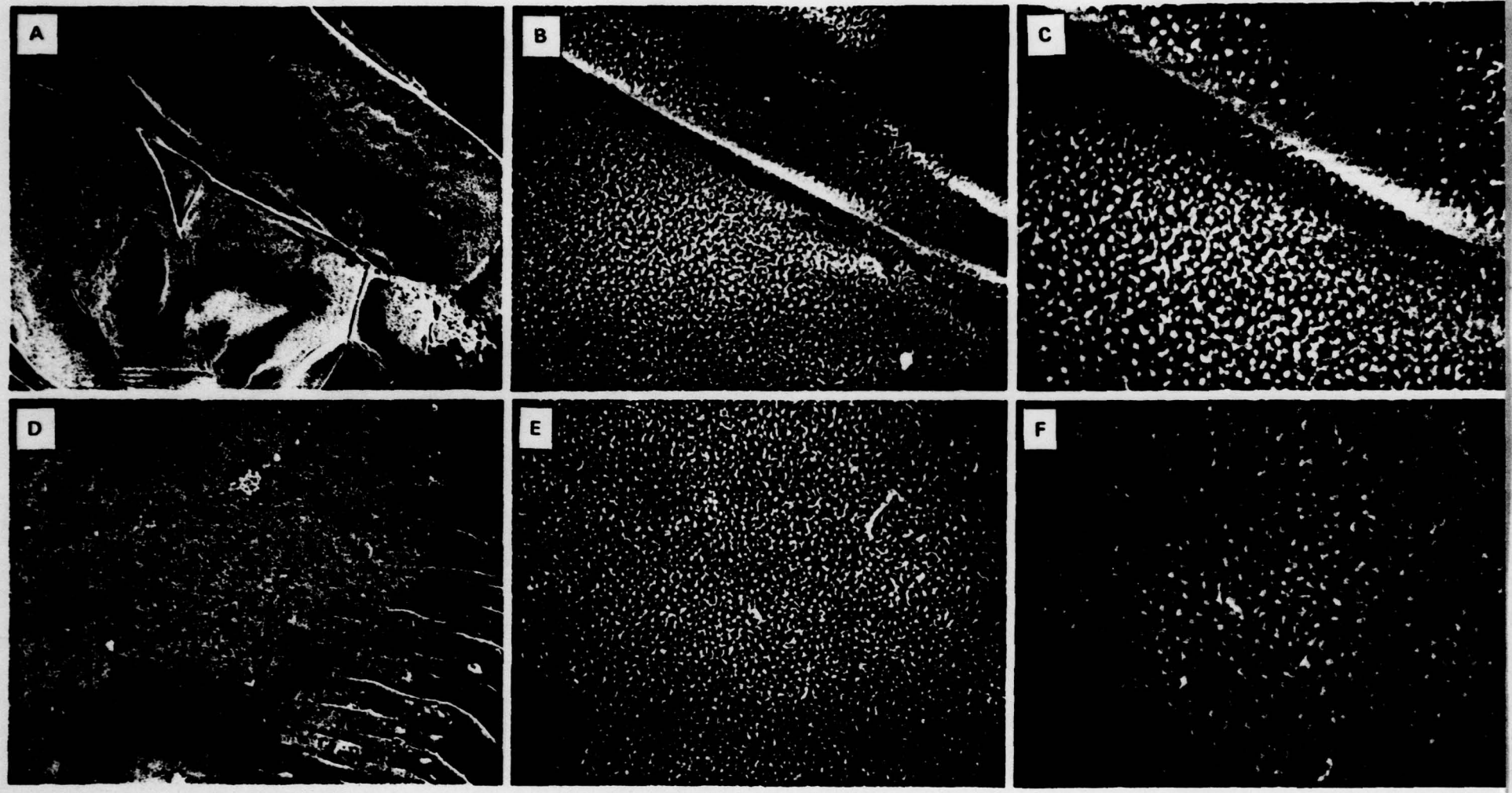

Figure B30. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 3 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

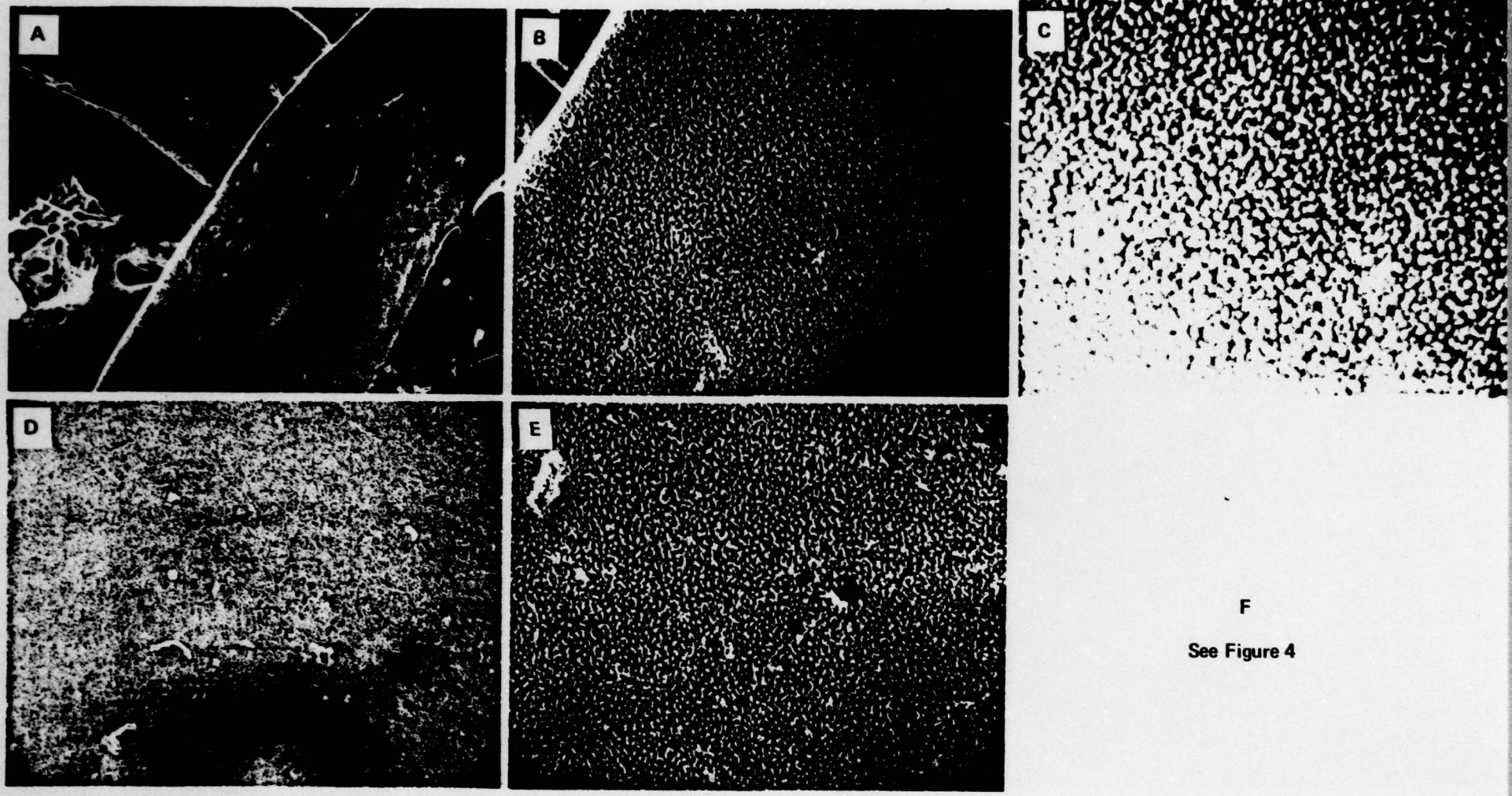

Figure B31. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 5 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

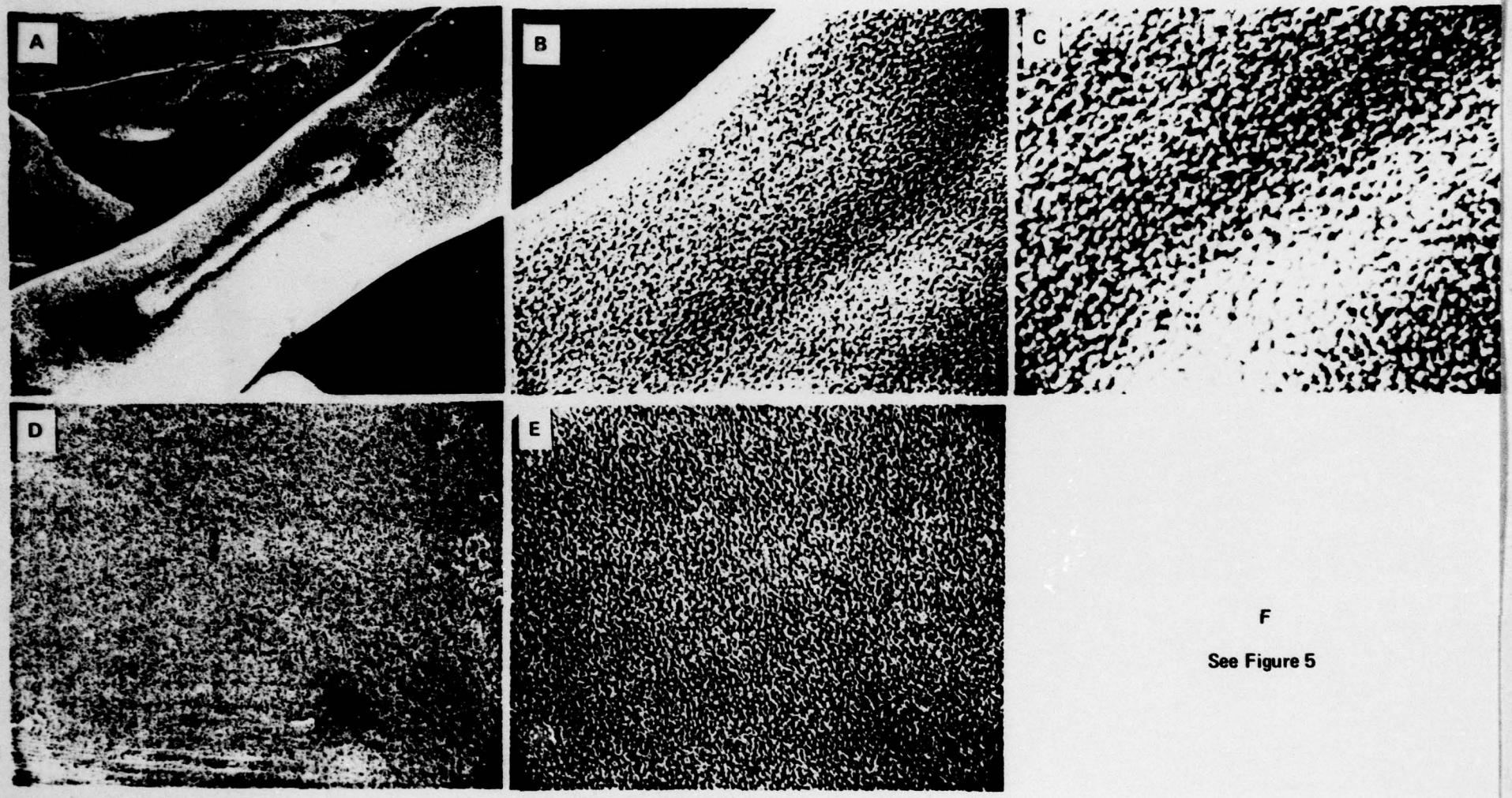

Figure B32. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 45 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

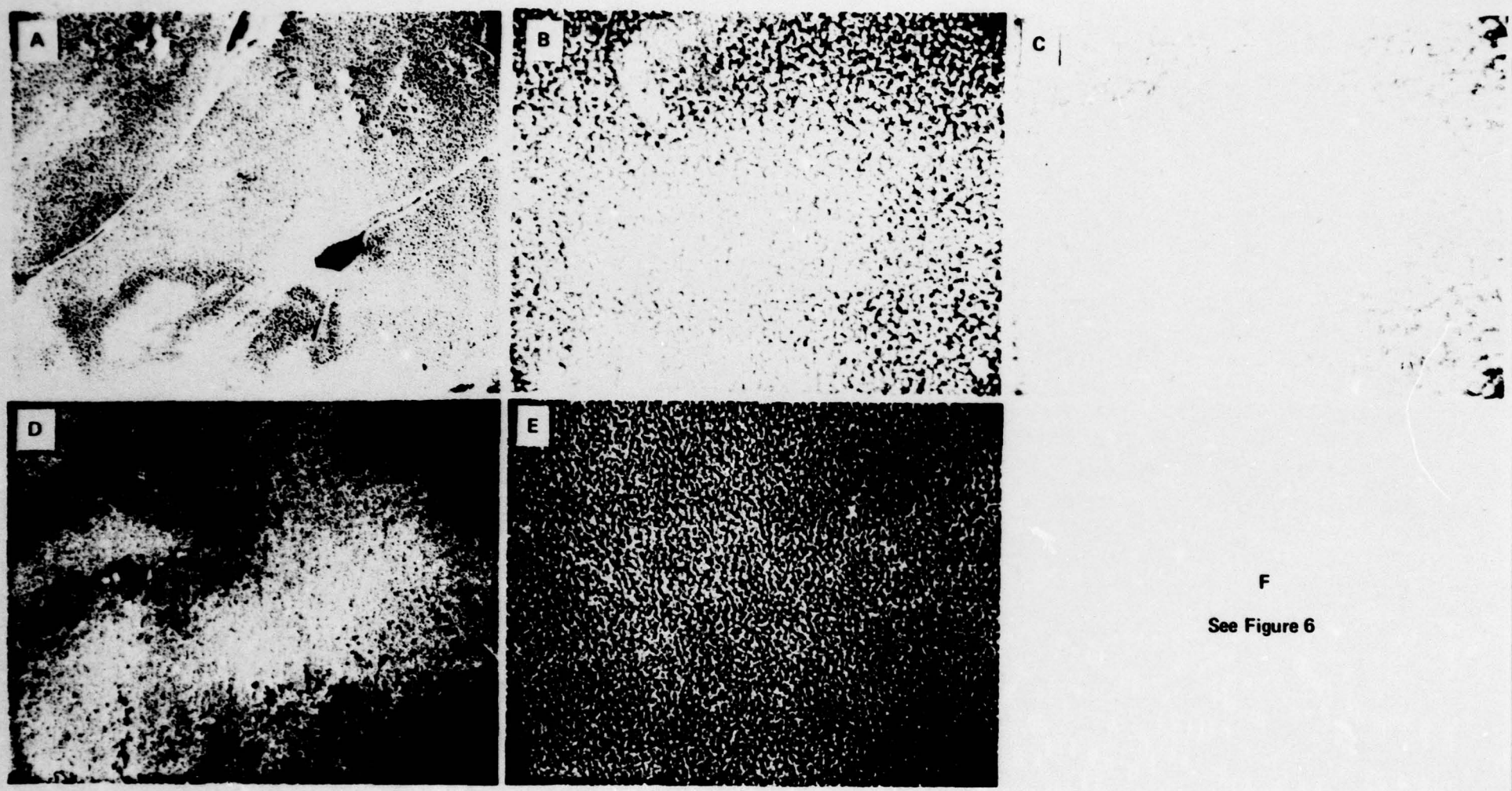

Figure B33. Scanning Electron Micrographs of the Front and Back of a Polyamide Membrane After Immersion
Immersion Time = 83 Days; Iodine Concentration = 50 ppm
Membrane Backs = A – 1,400X; B – 7,000X; C – 14,000X
Membrane Fronts = D – 1,400X; E – 7,000X; F – 14,000X

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