

BIOFOULING AND CORROSION FOULING OF PLAIN AND ENHANCED ALUMINUM SURFACES *

C. Panchal
Argonne National Laboratory
Argonne, IL 60439

D. Sasscer
Northwestern University
Evanston, IL 60208

ABSTRACT

A long-term experimental research was carried out for the qualification of aluminum alloys for marine applications as a part of the Ocean Thermal Energy Conversion (OTEC) program. This paper presents the short- and long-term biofouling and corrosion-fouling results for a rectangular-flow channel and for the spirally fluted tube, and compares these results with that for a plain tube. The rectangular-flow channel tested in the present experimental program represents a typical flow passage for brazed-aluminum heat exchangers. The effectiveness of biofouling control methods such as chlorination and brush cleaning were also evaluated.

INTRODUCTION

Corrosion is very important consideration in the design and development of heat exchangers. Although heat exchangers can be built with corrosion-resistant materials, they are generally expensive and they may not be suitable for advanced heat-exchanger configurations. Another very important consideration for the design of heat exchangers is fouling. In applications where heat exchangers must operate with small mean-temperature differences, it is essential to use high-performance heat exchangers with enhanced surfaces. For OTEC power systems, it was critical to develop high-performance and low-cost heat exchangers for the improvement of the overall system efficiency and to make the plant cost competitive. Aluminum alloys, due to their desirable mechanical properties, provide an opportunity to develop enhanced and compact heat-exchange surfaces. However, aluminum alloys are susceptible to corrosion; therefore, an experimental investigation was carried out to determine the corrosion behavior of selected aluminum alloys in tropical seawater. The following geometries were investigated: a plain tube, a spirally fluted tube, and a rectangular-flow channel.

It is well known that when engineering materials are exposed to natural waters, growth of biofilm occurs and its development consists of four sequential events; 1) adsorption of natural polymers, 2) attachment of microorganisms, 3) establishment of primary biofilm, and 4) deposition and growth of other microorganisms (Corpe, 1979; Kent, 1988). For aluminum alloys, the biofilm growth and the deposition of the corrosion products occur in parallel.

Very little is known about the interactive effects of the simultaneous growth of the biofilm and the corrosion film. Which of the four biofouling events is affected by the presence of corrosion film is not understood. It was reported (Pinheiro et al., 1988; Gerchakov et al., 1977) that certain metals inhibit the initial biofilm growth. However, the inhibition effects of certain metal surfaces diminish with time, and the biofilm growth becomes independent of the metal surface. On the other hand, how the diffusion of oxygen to the metal surface and the dissolution of corrosion products are influenced by the biofilm are not known. These phenomena are further complicated by the complex local-flow field in the vicinity of the wall surface for enhanced and non-circular flow channels. In the present investigation, the experimental results are provided which reveal some of the basic interactive effects of biofouling and corrosion fouling of aluminum alloys for plain and enhanced tubes and for non-circular flow channels.

EXPERIMENTAL ANALYSIS

The experiments were carried at the Natural Energy Laboratory of Hawaii with the tropical-surface and deep-ocean cold seawater. The typical water parameters are summarized in Table 1. Note the difference in dissolved oxygen, pH, inorganic and organic constituents for warm and cold seawater. Only the warm water results are discussed because there was no detectable biofouling of test sections exposed to the deep-ocean cold seawater.

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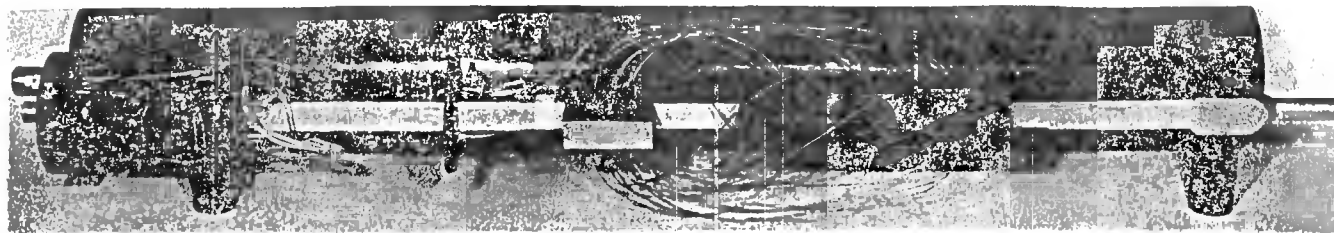


Figure 1. The Heat Transfer Monitor (HTM) Installed on the Rectangular-Flow Channel

Table 1. Comparison of Physical and Chemical Parameters for Seawater

	Warm Water (100 m offshore)	Cold Water (900 m deep)
Salinity, mg%	34.7	34.3
pH	8.1	7.54
Constituents, mg-atom/l		
PO ₄	0.14	2.97
NO ₃ + NO ₂	0.15	40.1
Si	3.34	77.7
Dissolved Oxygen, ml/l	5.00	1.05
Temperature, °C	28.9 ¹ 24.8 ²	8.5 - 10.5
Total Organic Carbon, mg/l	0.75	0.38

1 summer
2 winter

The fouling resistance values were measured with a Heat Transfer Monitor (HTM) as described by Panchal et al. (1991). An overall view of the HTM device installed on the rectangular-flow channel is shown in Figure 1. Short sample sections (about 100 to 150 mm long) were also installed down stream of the HTM and they were periodically withdrawn for chemical analysis of the fouling film.

Selected commercial grade aluminum alloys (Table 2) were investigated to determine their corrosion behavior in seawater. Figure 2 shows an elemental section of the brazed-aluminum heat exchanger for which the rectangular channel was tested. Note that water channels were prepared by an extrusion process. The seawater surface was coated with zinc applied by using a diffused-zinc technique to protect it from possible pitting corrosion. The spirally fluted-tube test section shown in Figure 3 was manufactured from extruded Al-6061 strips by biased-wrapping and welding along the seams. The plain-tube test sections were prepared from standard commercial tubes.

FOULING RESULTS

As mentioned earlier, the fouling results for only the warm seawater are presented in this paper. First, the short-term biofouling results are presented, followed by the results dealing with the effectiveness of chlorination for biofouling control. Finally, the long-term fouling results are presented, which show the combined effects of biofouling and corrosion fouling.

Table 2. Aluminum Test Sections

Test Sections	Aluminum Alloys	Manufacturing Process	Surface
Plain Tube	Al-3003	Seamless Drawn	Bare
Plain Tube	Al-5052	Seamless Drawn	Bare
Fluted Tube	Al-6061	Spirally Welded	Bare
Rectangular-Channel	Al-3003	Extrusion	Diffused-Zinc

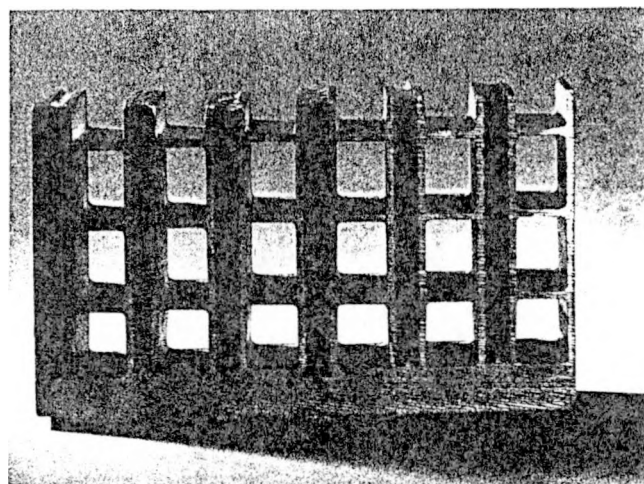


Figure 2. Elemental Section of the Compact Heat Exchanger Showing Extruded Water Flow Channels

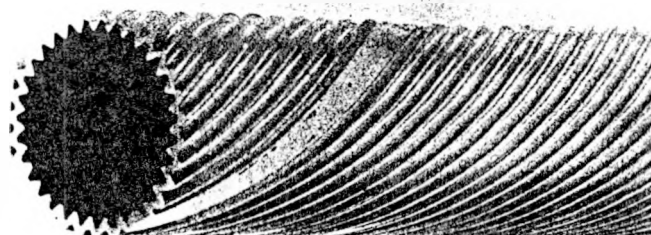


Figure 3. An Overall View of the Spirally Fluted Tube

Short-Term Biofouling Data

It was observed that it took about 200 days for a corrosion film to fully develop in warm seawater. After this initial period of rapid corrosion, the rate of corrosion reached an asymptotic value (Panchal et al., 1991). It will be shown later that the corrosion fouling resistance was less than about $0.008 \text{ m}^2 \text{ K/kW}$ for the first 30 days of testing. Therefore, the initial fouling-rate data for the first 30 days could be assumed to be mostly due to the biofilm growth. Figure 4 shows these initial fouling data. Note that the biofouling rates for the spirally fluted tube and for the rectangular-flow channel were comparable with that of the Al-5052 plain tube.

Also shown in Figure 4 are the fouling rates for a plain and for the spirally indented tube (Korodense LPD) made with a corrosion-resistant AL-6X stainless steel, which were reported previously by Panchal (1989). The results clearly indicated that the initial rate of biofouling was insensitive to the enhanced surface geometry and were similar for aluminum and AL-6X stainless steel.

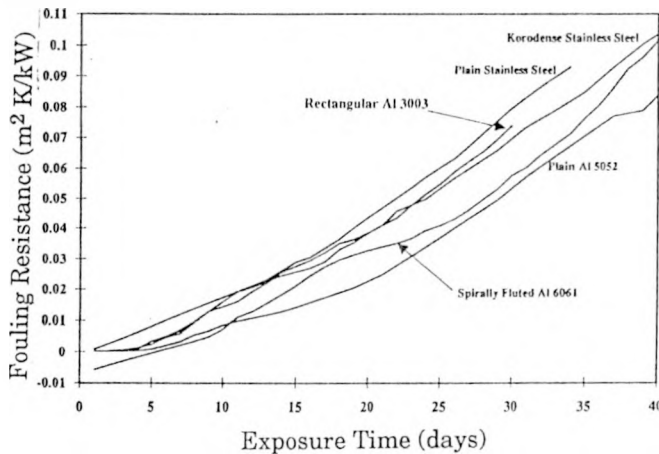


Figure 4. Comparison of the Rate of Biofouling in Warm Seawater for Plain and Enhanced Surfaces of Different Materials -- Stainless Steel and Aluminum Alloys

Chlorination Control for Plain Tubes

Both biofouling and corrosion fouling occur simultaneously in the surface warm water. Panchal et al. (1984) showed that for a corrosion-resistant material (e.g., titanium), daily chlorination of 70 ppb for one hour maintained the biofouling resistance value less than $0.009 \text{ m}^2 \text{ K/kW}$ ($0.00005 \text{ hr ft}^2 \text{ }^\circ\text{F/Btu}$). The technical issue to be resolved was the level of chlorination required for aluminum alloys to maintain the same biofouling resistance. To resolve this issue, a selected number of biofouling samples were analyzed to determine the effectiveness of chlorination for aluminum alloys.

In the initial phase of the Al-5052 experiments with the plain-tube test section, biofouling was allowed to occur without any biofouling control. Then, daily chlorination of 100 ppb for one hour was employed until the fouling resistance reached a low value and further chlorination would not reduce it. Three such uncontrolled fouling excursions followed by chlorination cycles were carried out as shown in Figure 5. After completing this initial experimental phase, this Al-5052 test section was chlorinated daily with 100 ppb for one hour. For the Al-3003 plain-tube test section, the chlorination schedule of 100 ppb for one hour per day was started from the beginning of the experiment as shown in Figure 6.

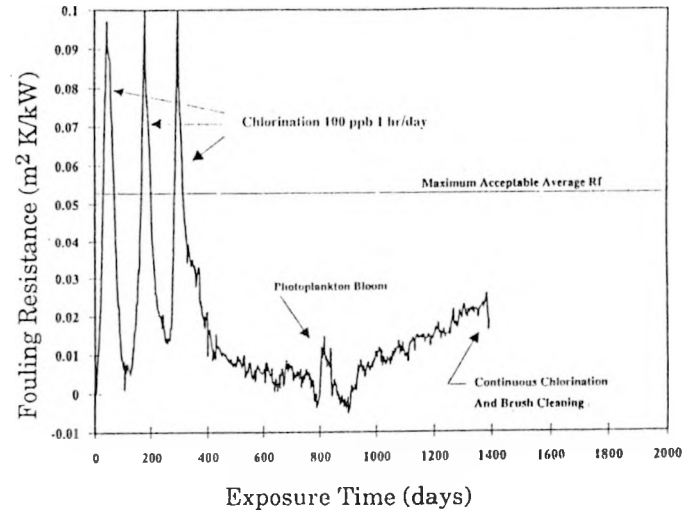


Figure 5. Fouling Data for Al-5052 Plain Tube; After 400 Days, Biofouling Controlled by Chlorination of 100 ppb One Hour per Day

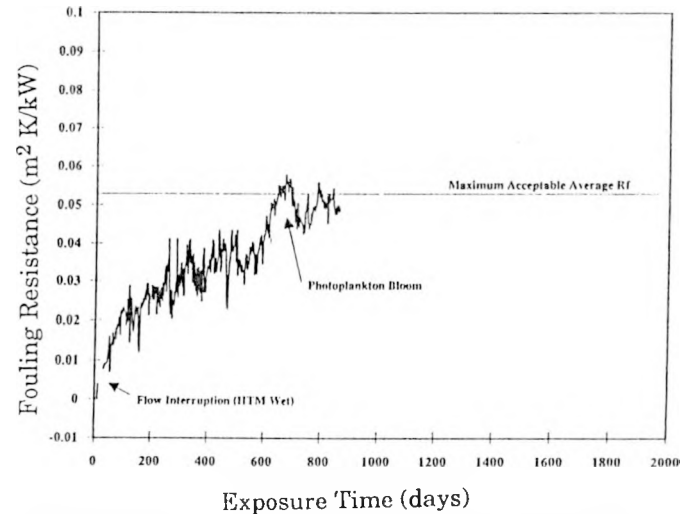


Figure 6. Fouling Data for Al-3003 Plain Tube; Biofouling Controlled by Chlorination of 100 ppb One Hour per Day

During the test period, short sample sections were withdrawn and the fouling film was analyzed in the laboratory. Details of the film analysis are given by Berger and Berger (1986). Tables 3 and 4 present the biofilm results for the Al-5052 and Al-3003 alloys, respectively. Due to inherent difficulties of removing the fouling film, this analysis was not done for the enhanced sections. Typical film analysis included measurements of the total fouling film (dried) weight, the organic nitrogen and carbon, and the physical examination by taking Scanning Electron Microscope (SEM) photograph of the fouling film. Note that the dried-film weight included both the biofilm and corrosion products.

The results can be summarized as follows:

- o For plain titanium tubes, the fouling resistance was less than $0.0035 \text{ m}^2 \text{ K/kW}$ for organic (carbon + nitrogen) matter weight of about $20 \text{ } \mu\text{g/cm}^2$ (Berger and Berger, 1986).

Table 3. Biofouling Film Analysis for Al-5052 in Warm Seawater, Uncotrolled Biofouling Followed by Chlorination of 100 ppb for One Hour per day Starting from and Ending on the day as Shown

Test Period (d)	R_f (m^2K/kW)	Dry Weight ($\mu g/cm^2$)	Nitrogen(N) ($\mu g/cm^2$)	Carbon(C) ($\mu g/cm^2$)	(N + C) ($\mu g/cm^2$)	(N + C)/Dry
177	0.105	217.1	4.51	18.38	22.89	0.11
180 ¹						
205	0.040	271.5	7.40	28.10	35.50	0.13
212	0.033	119.8	3.57	11.91	15.48	0.13
250	0.014	115.2	3.71	12.44	16.15	0.14
272	0.017	316.0	3.31	14.36	17.67	0.06
273 ²						
297	0.094	372.9	10.21	44.55	54.76	0.15
298 ¹						
320	0.046	275.0	4.15	16.14	20.29	0.07
372	0.032	172.6	4.38	16.56	20.94	0.12

1 Chlorination Started

2 Chlorination Ended

Table 4 Biofouling Film Analysis for Al-5052 in Cold Water, Uncotrolled Biofouling

Test Period (d)	R_f (m^2K/kW)	Dry Weight ($\mu g/cm^2$)	Nitrogen(N) ($\mu g/cm^2$)	Carbon(C) ($\mu g/cm^2$)	(N + C) ($\mu g/cm^2$)	(N + C)/Dry
31	0.008	808.0	0.27	3.85	4.12	0.01
59	0.013	585.8	1.52	4.56	6.07	0.01
91	0.022	422.0	1.61	4.94	6.55	0.02
121	0.029	638.8	1.24	7.02	8.27	0.01
150	0.021	451.6	0.85	6.39	7.24	0.02
182	0.029	430.3	0.55	4.03	4.58	0.01

- o During the initial experiment for Al-5052 test section, the corrosion film buildup continued, which can be seen in Table 3 at low values of organic matter.
- o Chlorination of 100 ppb applied one hour per day from the start for Al-3003 in warm seawater kept the biofilm growth to the minimum as indicated by the low values for organic matter shown in Table 4.
- o During the first 30 days of testing, Table 4 shows that the corrosion-fouling resistance was less than about 0.008 $m^2 K/kW$.
- o When the biofouling resistance was small, the fraction of organic carbon + nitrogen was in the range of 2 to 5% of the dry-film weight.

In the deep-ocean cold water, the biofilm growth for corrosion-resistant materials was negligibly small yielding undetectable fouling-resistance values (Panchal et al., 1984). For aluminum alloys deposition of organic (carbon + nitrogen) matter was less than $10 \mu g/cm^2$. As a result, the observed fouling resistance for aluminum alloys in the cold seawater could be assumed to be mainly due to the in-situ corrosion products.

Long-Term Fouling

The measured fouling resistance during the initial 30 days could be attributed to the biofilm growth; however, corrosion fouling became a significant part of the

measured fouling resistance for the extended time of these experiments. The biofilm buildup for the spirally fluted tube and for the rectangular-flow channel were controlled by daily chlorination of 70 ppb for one hour, which was found to be marginal for controlling the biofilm growth for the latter. For the plain tube sections (Al-5052 and Al-3003), higher levels of 100 ppb of chlorination applied for one hour was effective for keeping the biofilm growth to the minimum.

Figures 5 to 8 show the fouling-resistance variations with time obtained with the HTM's for the entire period of the experimental program for all four test sections: the two plain tubes, the spirally fluted tube, and the rectangular-flow channel.

Figure 5 shows the fouling-resistance variation for the Al-5052 test section. The fouling results showed some unusual behavior after completion of the initial fouling experiment or after about 400 days. The fouling resistance dropped to about $0.005 m^2 K/kW$ and stayed constant for an extended period of time. Moreover, it dropped to nearly zero, which was believed to be due to the measurement errors at both 800 and 900 days. There was a sudden rise in the fouling resistance at about 800 days which was attributed to an unusual water quality change -- a photoplankton bloom -- which lasted for a few days. This phenomena was observed with all four test sections. After 950 days, a normal corrosion-fouling pattern appeared to develop. At the conclusion of experiment (about 1400 days), the test section was chlorinated continuously for 24 hours followed by brush cleaning. Note that the fouling resistance did not

significantly changed indicating the surface was relatively free of biofilm and corrosion fouling was dominant.

Figure 6 presents the results for the Al-3003 plain tube test section. These results show that the fouling resistance followed an exponential trend which was the expected pattern with the fast initial rate followed by a rate reduction due to the corrosion-film buildup (Somerscales, 1981). Also note the sharp increase of the fouling resistance observed at about 700 days due to a photoplankton bloom mentioned above. As shown in Table 4, the biofilm growth was relatively low; therefore, the measured fouling resistance was mostly due to the in-situ corrosion products.

The fouling-resistance variation for the spirally fluted tube is shown in Figure 7. After continuous chlorination (about 100 days into the experiment), a gradual buildup of corrosion fouling occurred, but the fouling resistance suddenly increased at about 200 days due to a photoplankton bloom mentioned above. The fouling resistance was later reduced (about 550 days) by applying continuous chlorination for 16 hours. This was followed by a systematic increase in the fouling resistance until about 800 days when the experiment was terminated.

Figure 8 shows the fouling resistance variation of the rectangular test section. Daily chlorination level of 70 ppb applied for one hour could not effectively control the biofilm growth. The fouling trend showed the effect of seasonal variation in which the rate of fouling was higher during the summer than during the winter. At the end of the test period, continuous chlorination followed by nylon brushing reduced the fouling resistance from about 0.092 $\text{m}^2 \text{K/kW}$ to 0.029 $\text{m}^2 \text{K/kW}$.

It is interesting to note that except for the Al-3003 plain-tube test section, the interpreted value for the corrosion-fouling resistance was about 0.02 $\text{m}^2 \text{K/kW}$. The corresponding dried-film thickness for these alloys were in the range of 13 to 19 μm . Further, Panchal et al. (1991) reported that the thermal resistance of the dried film was about half the value for the hydrated corrosion film.

DISCUSSION OF INTERACTIVE EFFECTS

Interactive Effects of Biofouling and Corrosion Fouling

This experimental program did not investigate the interactive effects of biofouling and corrosion fouling exclusively for extended operating periods. Nevertheless, it was possible to make several overall observations. The first was that the corrosion rate was not influenced by the presence of biofilm on aluminum surfaces. Panchal et al. (1991) reported that the rate of corrosion was independent of the two biofouling control methods: 1) chlorination, and 2) uncontrolled fouling followed by manual brushing. The biofilm thickness for these two control methods were different during the test period. Therefore, it can be concluded that the diffusion of oxygen to the metal surface was not significantly affected by the presence of biofilm and the corrosion rate was not dependent on the biofouling history. The second observation was that the initial biofilm growth was comparable to that for AL-6X stainless steel, which indicates that aluminum surface did not inhibit the deposition of organisms and subsequent biofilm growth. A higher concentration of chlorination (100 ppb) was required for maintaining the biofilm growth comparable to that for corrosion-resistant materials suggests that the biofilm got some protection from the corrosion film, either against the oxidizing effects of chlorine or against subsequent removal of the oxidized biofilm.

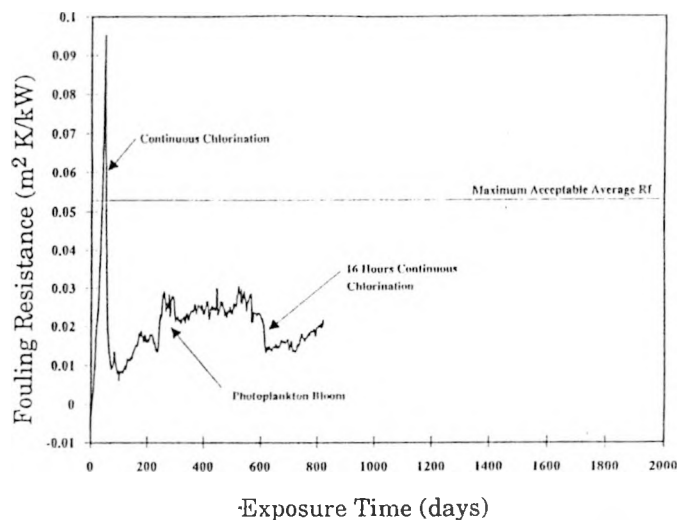


Figure 7. Fouling Data for the Al-6061 Spirally Fluted Tube; After 70 Days, Biofouling Controlled by Chlorination of 70 ppb One Hour per Day

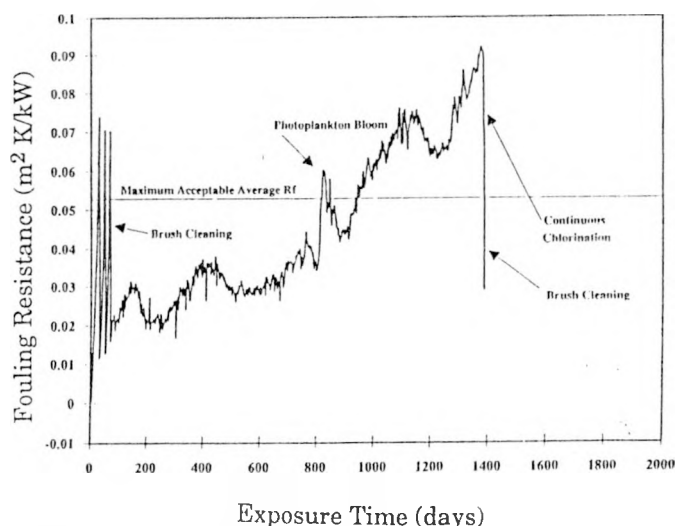


Figure 8. Fouling Data for the Diffused Zinc Al-3003 Rectangular-Flow Channel in Warm Seawater, After 70 Days, Biofouling Controlled by Chlorination of 70 ppb One Hour per Day

Interactive Effects of Fouling and Enhancement

Very little is known about the interactive effects of fouling and the flow field in the vicinity of the wall surface of an enhanced surface that provide the heat-transfer enhancement. The complex nature of distribution of shear stress and turbulence levels could lead to localized development of the fouling film and it could influence the effectiveness of the biofouling control method(s).

Corrosion Fouling. It is generally assumed that the rate of corrosion is inversely proportional to the oxygen-diffusion barrier of the corrosion-product film (Somerscales, 1981). The results from the present investigation showed that the long-term rate of corrosion fouling was not strongly dependent on the local-flow field of an enhanced and non-circular flow channels. In addition, removal of the corrosion products by the dissolution method could be affected by the local-flow field of enhanced and non-circular channels. But the

dissolution process may be slow (Dillon, 1959), and no significant effects of the local-flow field on the rate of removal of corrosion film could be expected. However, it should be noted that the presence of solid particulates was minimal in the present study and the fluid velocity was in the range of 1.4 to 1.8 m/s. Therefore, care should be taken for applying the same conclusion for flow conditions where the fluid velocity and particulate concentration are relatively high.

Biofouling. The short-term biofouling data did not show a strong effect of the enhancement geometry or passage shape on either the initial rate of biofouling or the long-term corrosion fouling rates. Therefore, it could be concluded that very weak fouling and enhancement interactions existed for these seawater experiments. The initial biofilm growth rates for the spirally fluted tube and for the rectangular-flow channels were comparable to that for plain tubes.

Chlorination Effectiveness. It should be noted that the surface and passage geometry did have some influence on the effectiveness of chlorination to remove the biofilm. For example, the effectiveness of 70 ppb chlorination for the spirally fluted tube shown in Figure 6 was comparable to that for titanium (Panchal et al., 1985) for maintaining the fouling resistance below 0.02 m² K/kW. This finding suggests that the swirl-induced mechanism promoted removal of the oxidized biofilm. It was also shown that the chlorination effectiveness was reduced for the rectangular-flow channel. The enhanced and rectangular flow channels would create flow field with non-uniform shear-stress and turbulence-intensity distributions. The effectiveness of chlorination could be significantly reduced at surface locations where the local turbulence intensity is lower than that for plain surfaces.

The process of removal of biofilm by chlorination consists of two major steps: 1) oxidation of biofilm by chlorine (or other oxidant like ozone), and 2) removal of oxidized biofilm from the surface. There are two possible mechanisms for removal of the oxidized biofilm. The lift force created by the velocity gradient in the boundary sublayer can dislodge the biofilm and remove it from the surface. Alternatively the random instantaneous turbulence burst can breakup the biofilm and lift it from the surface. Note that 70 ppb was effective for controlling the biofilm growth for the spirally fluted tube, but the same level of chlorination could not control the biofilm growth effectively for the rectangular-flow channel probably due to the ineffective removal of the biofilm from corners. The chlorination schedule of 100 ppb applied for one hour per day was effective for controlling the biofilm growth for both aluminum plain-tube sections. No definite conclusion could be derived from the overall results discussed above. A detailed analysis is required to compare the rate of diffusion, the kinetics of oxidation, and the rate of removal of the biofilm to determine the controlling mechanism. An analysis similar to that carried out by Characklis (1980) for a corrosion-resistant material in a laboratory environment should provide quantitative data for characterizing the mechanism of biofilm removal from aluminum surfaces.

Slightly higher level of chlorination was required for controlling the biofilm growth on aluminum alloys than on titanium. Therefore, it can be stated that the corrosion film either diminishes the oxidizing action of chlorine or that it protects the oxidized biofilm from being dislodged from the surface by the local-flow field.

CONCLUSIONS

The major conclusions derived to characterize fouling of plain surfaces, enhanced surfaces and non-circular flow channels are as follows:

- o The initial rate of biofouling was essentially independent of the enhancement geometry, the channel geometry, and the material type (aluminum Al-6061, Al-3003 and Al-5052, and AL-6X stainless steel).
- o The effectiveness of chlorination for the rectangular-flow channel was slightly less than that for plain tubes.
- o The interactions between biofouling and corrosion fouling appeared to be insignificant.
- o The rate of corrosion fouling was relatively insensitive to the enhanced tube and to the rectangular-flow channel. This finding supports the hypothesis that the rate of corrosion is controlled by the diffusion of oxygen through the corrosion film and it is unaffected by the local-flow field.

Additional conclusions derived from this long-term research program for qualification of aluminum alloys for marine applications are included in the report by Panchal et al. (1991).

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