

## ON-LINE MONITORING AND QUANTITATIVE ANALYSIS OF BIOFOULING IN LOW-VELOCITY COOLING WATER SYSTEM

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**Abstract** – To monitor and analyze the biofouling phenomenon caused primarily by microbial suspended solids (MSS), a 'biofouling tube' apparatus consisting of a carbon steel tube, a differential pressure transmitter, a corrotor, and a cooling water circulation device was designed and fabricated. By measuring continuously the pressure drop across the biofouling tube and using the correlation between the fluid's friction factor and surface roughness, we quantitatively analyzed the influence of MSS concentration, temperature, and fluid velocity on the rate of biofilm growth on the metal surface. The result indicated that the fluid velocity had the most profound impact, e.g., lowering the linear velocity from 0.3 to 0.15 m/sec resulted in about a four times higher biofouling rate. Up to 50 ppm MSS, the biofouling rate was proportional to the MSS concentration. The biofouling rate at 35 °C was about 1.75 times higher than that of 45 °C, probably due to the diminishing effect of thermolabile microorganisms exposed at 45 °C. It was also demonstrated that the biofouling could be significantly reduced by incorporating cooling water treatment programs such as protective pre-filming and adding corrosion inhibitors. This apparatus, if installed on-site at a sidestream of a cooling water system, could be used to monitor the biofouling tendency on-line and to suggest timely preventive maintenance actions.

**Key words:** Biofouling, Microbial Suspended Solids, Pitting, Corrosion, Cooling Water Treatment, Biofouling Monitoring

### INTRODUCTION

Nearly all the chemical processing industries use heat exchangers for heating, cooling, evaporation, concentration, etc. Among these, cooling is probably the most widespread application. Water is widely used as the cooling medium because it is relatively inexpensive and abundant. Recently, however, due to such problems as water resources shortage, water pollution, and concentrated usage, heat exchangers using cooling water have suffered from accelerated fouling and corrosion problems [Cheremisinoff, 1995].

The major factors that can cause the corrosion of a heat exchanger surface include microbial suspended solids, corrosive ionic species, operating temperature, and circulation velocity of cooling water. Of these, microbial suspended solids (MSS) are known to play a critical role. Because of their self-aggregating and floc-forming properties, MSS easily adhere to the heat exchanger surfaces and gradually form a dense and fibrous layer called 'biofilm' [Geesey et al., 1994; Busscher and Weerkamp, 1987]. As the biofilm gets thicker, the friction of the cooling water flow increases resulting in an increased pressure drop. Also, heat exchanger efficiency can be dramatically reduced, which eventually causes unexpected shutdown for repair [Pope et al., 1984; Tatnall, 1981]. Inside the biofilm, sulfur-reducing bacteria can proliferate because of the low availability of dissolved oxygen, and as a result surface

pitting becomes accelerated [Hamilton, 1985; Marcus, 1995].

Due to continually changing operation conditions, the causes and effects of biofouling have not been clearly elucidated. Industries rely heavily on experience for predicting and solving this problem [Uchida et al., 1990; Thierry, 1987; Tatnall, 1981; Colturi and Kozelski, 1984]. Therefore, it is necessary to develop a method to monitor the progress of biofouling and to quantitatively evaluate its consequences. In our study, we have designed and fabricated such a monitoring device called a 'biofouling tube'. Using this, we quantified the effects of the important parameters such as MSS concentration, flow velocity, and temperature on biofilm growth and pitting tendency. Furthermore, the effects of prefilming and anti-corrosion treatment protocols were evaluated. Biofouling and the consequent corrosion problem can lead to undesirable shutdown and repair, which in turn can increase expenditures and result in a loss of productivity. Installed on-site next to a cooling water system, the biofouling tube could enable us to monitor the actual progress of the biofouling on-line and continuously. Combined with the implementation of proper and timely anti-corrosion programs, it would provide significant economic and environmental benefits by extending maintenance-free operation of heat exchangers.

### MATERIALS AND METHOD

#### 1. Materials

The MSS used in this experiment was collected from an actual cooling water system and supplied by Hansu, Inc. (Ansan,

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Korea). Local tap water was used as the cooling water after both M-alkalinity and hardness were adjusted to 200 ppm (as  $\text{CaCO}_3$ ) for standardization of the test water. The biofouling tube was made of STB35 carbon steel which is widely used as a heat exchanger material. For standardization of the experiment, each biofouling tube was pretreated for degreasing and protective film formation. For degreasing, Sweelin<sup>®</sup>M (consisting primarily of a non-ionic surfactant) and Kurizet<sup>®</sup>S-204 (consisting of polyphosphate salts and zinc salt) were used. For prefilming, Kurita<sup>®</sup>S-3700 (consisting of polyphosphate salts) and Kurita<sup>®</sup>S-6000 (consisting of zinc salt and thus reacting with phosphate salts to form a tight protective layer) were used. For an anti-corrosion treatment, Kurita<sup>®</sup>S-1030 (major ingredients were organophosphates, azole compounds, and polymeric dispersant), Kurita<sup>®</sup>S-6000 (an anti-corrosion aid), and Polycin A-496 (as a sanitizer and slime dispersant) were added to the cooling water. All the materials were donated by Hansu, Inc.

## 2. Design and Fabrication of Biofouling Tube

A single tube model was selected over the shell-and-tube, since in the shell-and-tube model the fluid flow patterns are significantly different depending on the location, and thus any mathematical estimation and physical measurement inevitably bring about large deviations [Sanatgar and Somerscales, 1991; Taborek et al., 1972; Epstein, 1983]. A carbon steel tube with 14 mm ID, 19 mm OD, and 126 cm length was used. The L/D ratio was about 90 to minimize the entrance and the exit effects [Nevers, 1991]. Other fittings were copper, stainless steel, or silicon material. To precisely measure the pressure drop across the biofouling tube, a differential pressure transmitter (Konics, model LD 301-D111-TD11-0110) was used. It could measure a 3–40 mm  $\text{H}_2\text{O}$  pressure drop (equivalent to  $3 \times 10^{-4}$  to  $4 \times 10^{-3}$  atm). To monitor the corrosion tendency, a corrotor (Rohrbach Cosasco Systems, model 9030) was installed on the downstream side of the tube. Signal outputs from the transmitter and the corrotor were inputted to a personal computer via analog-to-digital conversion for on-line data acquisition and display. Fig. 1 shows the schematic diagram of the biofouling tube and its peripherals [Walker and

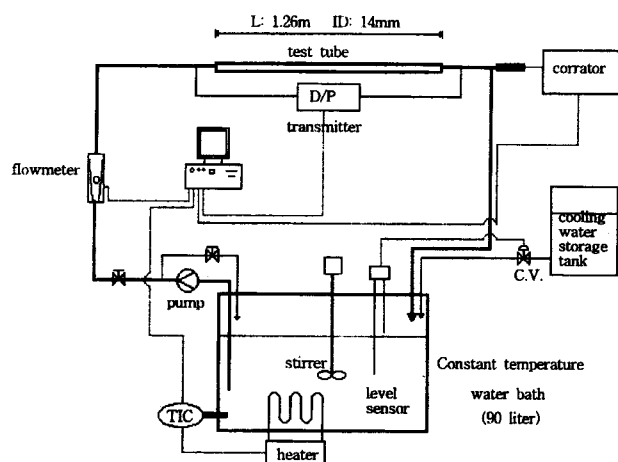


Fig. 1. Schematic diagram of biofouling tube and its peripherals.

Maddux, 1989; Wakamatsu et al., 1991].

## 3. Calculation of Biofilm Thickness from Pressure Drop Data

Considering that the biofilm growth would increase the surface roughness of the biofouling tube and using the correlation between friction factor and relative roughness in the conduit flow, we calculated the biofilm thickness from the pressure drop data. Friction factor ( $f_f$ ) can be related to the pressure drop ( $\Delta P$ ) as follows [Welty et al., 1984]:

$$f_f = \frac{\Delta P \cdot D}{2\rho L v^2} \quad (1)$$

where  $D$ ,  $\rho$ ,  $L$ , and  $v$  represent inside diameter, fluid density, tube length, and linear velocity of fluid flow, respectively. Also, the relationship of friction factor and relative roughness is expressed as follows [Welty et al., 1984]. In the transition flow regime,

$$\frac{1}{\sqrt{f_f}} = 4 \log_{10} \frac{D}{\epsilon} + 2.28 - 4 \log \left( 4.67 \frac{D \cdot \text{Re}}{\epsilon \sqrt{f_f}} + 1 \right) \quad (2)$$

In the turbulent flow regime,

$$\frac{1}{\sqrt{f_f}} = 4 \log_{10} \frac{D}{\epsilon} + 2.28 \quad (3)$$

where  $\epsilon/D$  and  $\text{Re}$  denote relative roughness of the inside surface of the tube and Reynolds number, respectively. Combining Eq. (1) with either Eq. (2) or (3), we could obtain  $\epsilon/D$  values from the pressure drop data. The  $\epsilon/D$  values thus obtained would give average thickness of the biofilm.

## 4. Experimental Procedure

Fig. 2 and Table 1 respectively show the experimental flow-

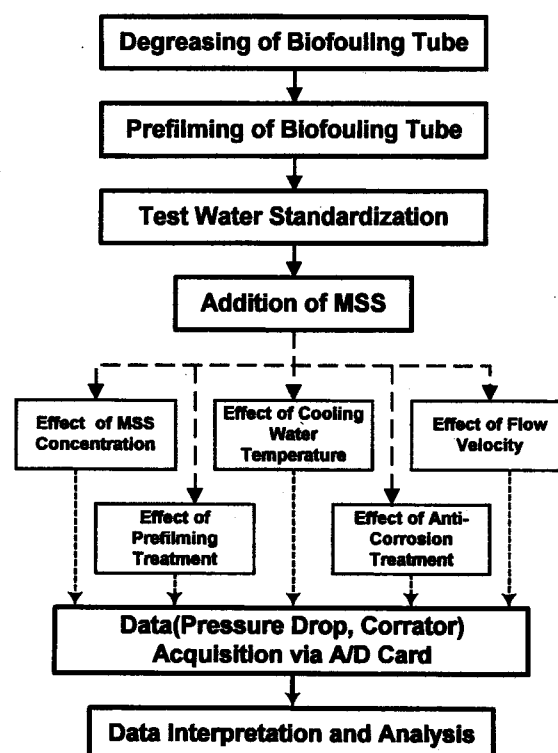


Fig. 2. Experimental flowchart.

**Table 1. Experimental conditions**

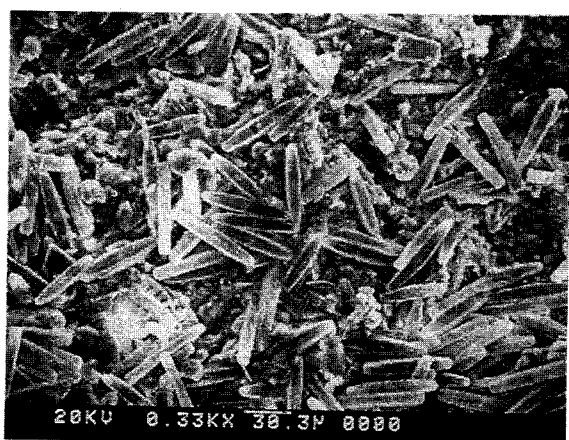
Experiment condition	Effect of MSS concentration	Effect of temperature	Effect of flow velocity	Effect of prefilming treatment	Effect of anti-corrosion treatment
Concentrations of MSS (ppm)	0, 10, 30, 50	50	50	50	50
Temperature (°C)	45	35, 45	35	45	35
Flow velocity (m/s)	0.15	0.15	0.15, 0.3	0.15	0.15

chart and the conditions used. In heat exchanger design, the tube side fluid velocity is usually set at 1.0-2.5 m/sec and the shell side velocity at 0.3-1.0 m/sec [Coulson et al., 1989]. In actual operations, however, cooling water is circulated at much lower flow rates. It is well known that the fouling and corrosion rates increase significantly at lower flow rates. In this experiment, two velocities were employed: 0.15 and 0.30 m/sec. MSS concentration is another important factor. Four different MSS concentrations (0, 10, 30, 50 ppm) were used in this experiment. Two temperatures (35 and 45 °C) were compared. The prefilming effect was evaluated at 45 °C, 50 ppm MSS concentration, and 0.15 m/sec velocity. The experiment to evaluate the performance of the anti-corrosion treatment was carried out at 35 °C, 50 ppm, and 0.15 m/sec. Cooling water pH was not controlled but it was maintained rather constant at 8.5-9.0 range. For the degreasing pretreatment, Sweelin<sup>®</sup>M and Kurizet<sup>®</sup>S-204 were added to the test water at 20-30 ppm and 200 ppm concentrations, respectively, and then circulated for 2 hours. For the prefilming treatment, Kurita<sup>®</sup>S-3700 and Kurita<sup>®</sup>S-6000 were added at 400 and 100 ppm at pH 6.5 and then circulated for 24 hours. For the anti-corrosion treatment, Kurita<sup>®</sup>S-6000, Polycin<sup>®</sup>A-496, and Kurita<sup>®</sup>S-1030 were added to the test water at 15, 100, and 80 ppm, respectively.

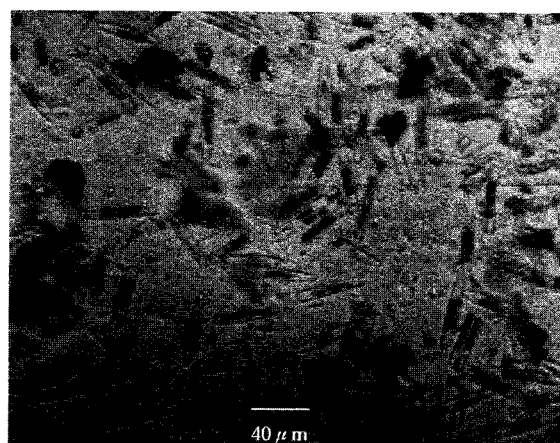
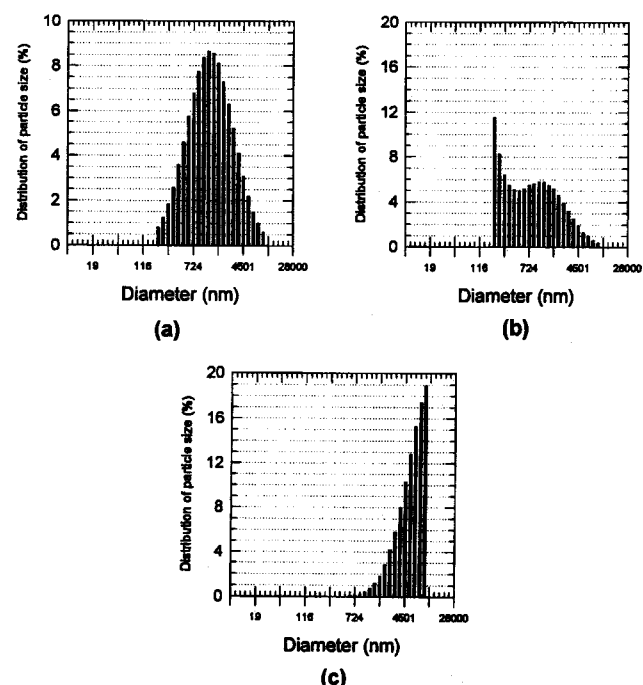
## RESULTS AND DISCUSSION

### 1. Characterization of MSS

Among the various kinds of microorganisms comprising the MSS, filamentous bacteria are known to possess the highest adherent property [Busscher and Weerkamp, 1987]. Photo 1(A) and 1(B) show the MSS used in this study. They indeed

**Photo 1(A). Scanning electron microscope of MSS.**

consisted of a variety of microorganisms, and several major species represented the microbial flora (or consortium). It was well supported by the size distribution data acquired from the DLS (dynamic light scattering) experiment (see Fig. 3.) The MSS had a normal size distribution ranging from approxi-

**Photo 1(B). Optical microscope of MSS.****Fig. 3. Size distribution data acquired from dynamic light scattering experiment.**

(a) Distribution of scattering intensity, (b) Number distribution of MSS, (c) Weight distribution of SS

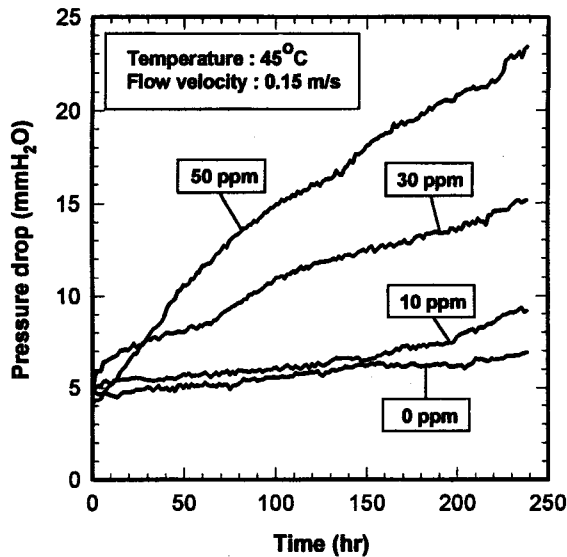


Fig. 4. Effect of MSS concentration on pressure drop.

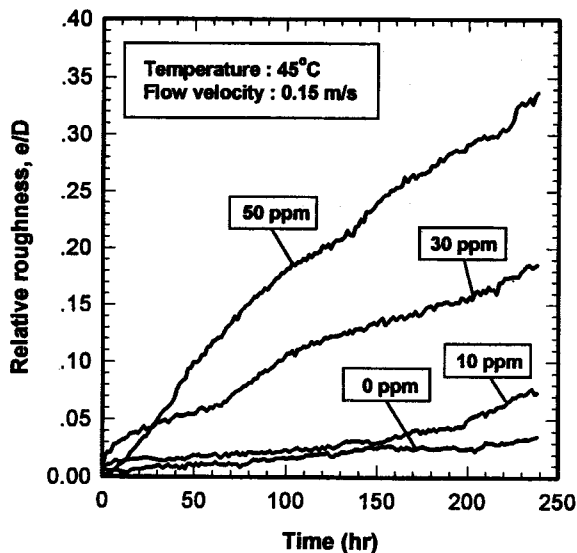


Fig. 5. Effect of MSS concentration on biofilm thickness.

mately 0.2  $\mu\text{m}$  to 10  $\mu\text{m}$  with an average diameter of ca. 1.2  $\mu\text{m}$ . The plating experiment showed the MSS had a viable cell density of  $2 \times 10^6$  CFU/ml. Solid content of the MSS was estimated at ca. 7%.

## 2. Effect of MSS Concentration on Biofouling

Fig. 4 shows that the pressure drop of the cooling water flow increased as the MSS concentration was increased. Also, the profile of the pressure drop increase was nearly linear with respect to time, indicating that the biofilm thickening process may be assumed to have zeroth order kinetics. Fig. 5 shows the plots of the changes in the  $e/D$ . As expected, both  $\Delta P$  and  $e/D$  changed very similarly. At 50 ppm MSS,  $e/D$  started to exceed 0.30 after about 200 hours of circulation. Also, at 50 ppm MSS or below, the biofouling rate increased linearly with time up to 10 days. The slopes calculated from the regressed profiles are shown in Fig. 6. This indicated the biofouling rate was proportional to the MSS concentration up to 50 ppm, and the biofilm growth rate ranged from

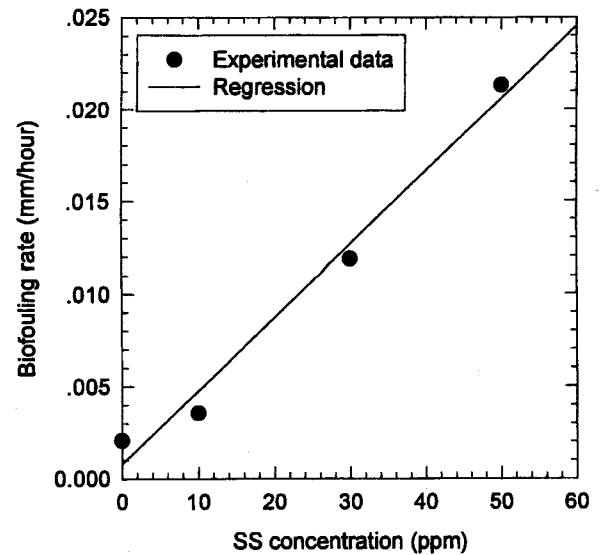


Fig. 6. Effect of MSS concentration on biofouling rate.

0.003 mm/hr at 10 ppm to 0.021 mm/hr at 50 ppm.

## 3. Effect of Temperature

The design temperature of the cooling water stream in heat exchanger design is usually 20–25°C, but in actual operations the cooling water temperature often exceeds 35°C. In this study, the temperature effect was studied at 35 and 45°C. As can be seen in Fig. 7, the biofouling rate at 35°C was about 1.75 times higher than that of 45°C, probably because the microorganisms present in the MSS grew better at 35°C. To confirm this, we cultured the MSS at the two temperatures and found that at 45°C the CFU density dropped to 1/100 after about 12 hours. However, yellow-colored colonies, probably some of the relatively thermostable cocci, continued proliferating as shown in Photo 2.

## 4. Effect of Flow Velocity

The design flow velocity for the cooling side fluid is usually around 0.3 m/sec; however, in actual operations cooling

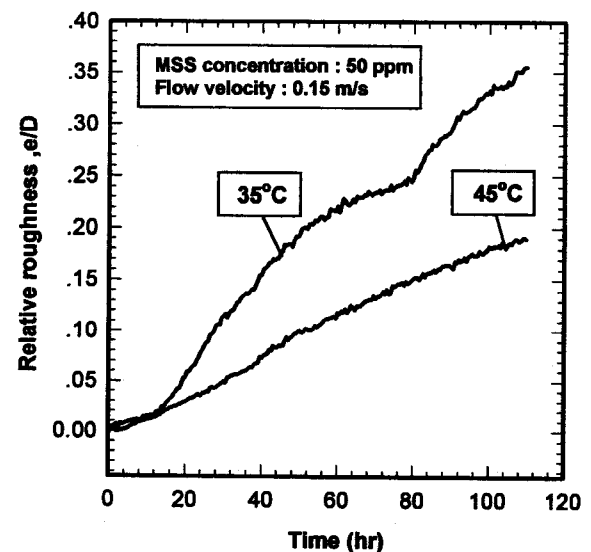


Fig. 7. Effect of temperature on biofilm thickness.

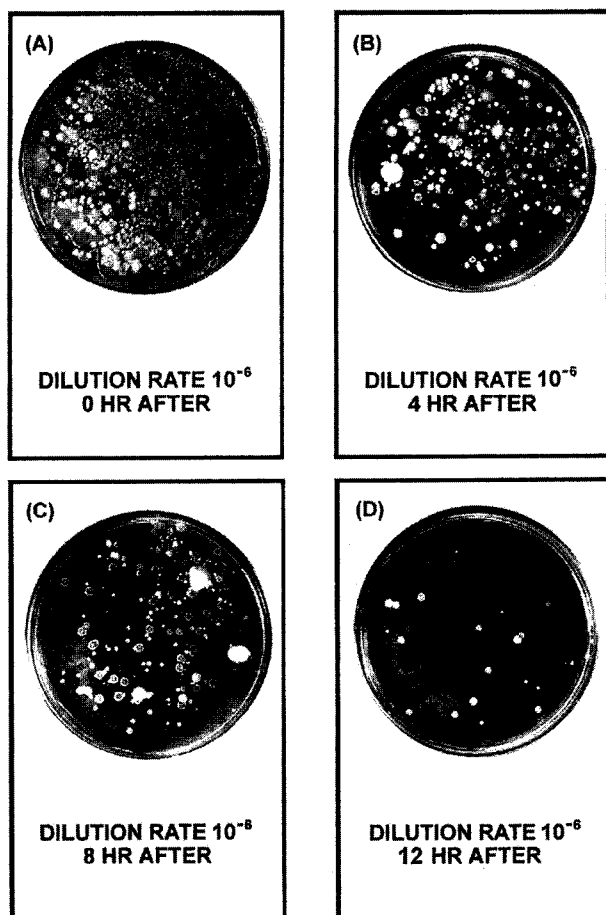


Photo 2. Agar plates of MSS cultured at 45 °C.

(A) initial, (B) after 4 hours, (C) after 8 hours, (D) after 12 hours

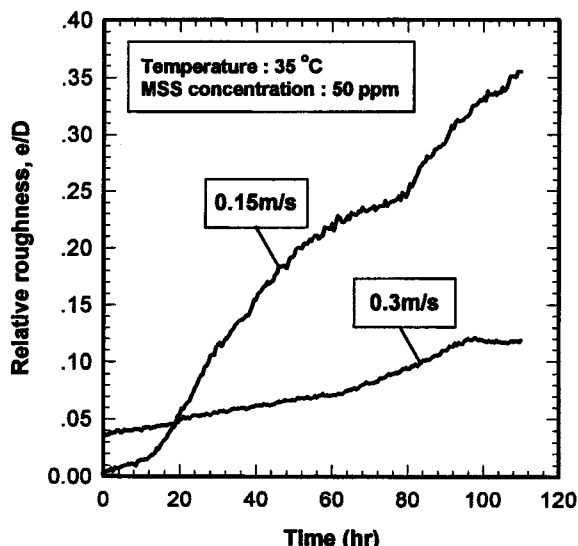


Fig. 8. Effect of flow velocity on biofilm thickness.

water is circulated at much lower velocities. In the low-velocity condition, the shearing effect is much reduced allowing increased rate of MSS adherence and growth on the surface. To evaluate this quantitatively, we compared the low (0.15

m/sec) and normal (0.3 m/sec) velocities. The result shown in Fig. 8 demonstrates that the biofouling rate at 0.15 m/sec condition was ca. 4 times higher than that at 0.3 m/sec. Thus, it can be concluded that among the factors studied the flow velocity is the single most critical parameter in controlling the biofouling.

#### 5. Effect of Prefilming Treatment

Two chemicals, i.e., Kurita®S-3700 and Kurita®S-6000, were added for prefilming (see "Materials and Methods" section for the concentrations and the procedure used). The former contains polyphosphates and reacts with divalent metal ions to form a film layer on the negative-charged metal surface, which in turn prevents the reduction of oxygen molecules. Kurita®S-6000 contains zinc salts which react with phosphates to form a layer and/or proceed to form a zinc hydroxide layer. Protective layers of a tight matrix nature can be formed by combining these two chemicals. As can be seen in Fig. 9, the biofouling rate could be suppressed by about 60 % by the prefilming treatment.

#### 6. Effect of Anti-Corrosion Additives

To maintain the cooling water quality, various chemicals are periodically added to suppress MSS proliferation, to minimize pitting by MSS flocculation, and to prevent scaling. Of the additives used, Kurita®S-1030 forms a  $\gamma\text{-Fe}_2\text{O}_3$  layer to prevent corrosion as well as to disperse SS and scale. Kurita®S-6000 acts as an aid to Kurita®S-1030 by forming a protective layer. Finally, Polycrin A-496 works on the slime layer to reduce its adhesivity, to help its detachment from the surface, and eventually to help disperse the detached slimes. It also has a sanitizing effect against bacteria and algae. The effect of such anti-corrosion additives was quantitatively evaluated. The result in Fig. 10 suggested that such additives could suppress the biofouling rate by about 90 %, particularly in the low-velocity condition.

#### 7. Simulation Study for Corrosion Measurement

Using the corrotor installed in the biofouling tube, we measured the corrosivity exerted by ionic species under given conditions. Fig. 11 shows that the measured corrosivity, like the

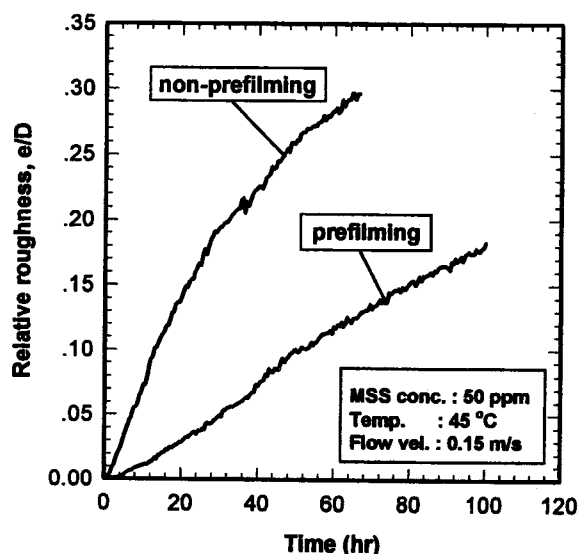


Fig. 9. Effect of prefilming on biofilm thickness.

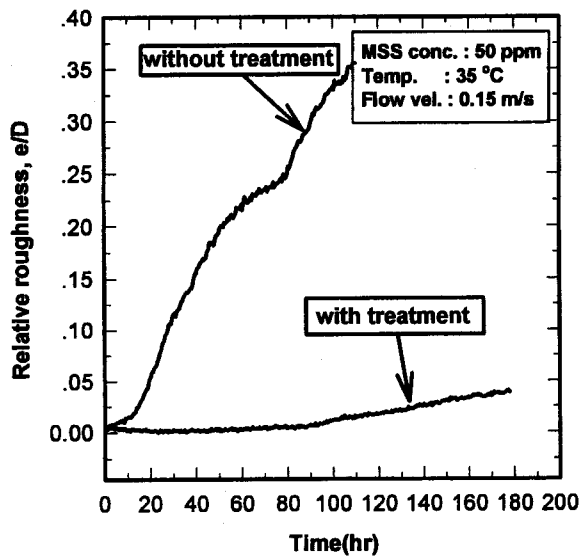


Fig. 10. Effect of anti-corrosion treatment on biofilm thickness.

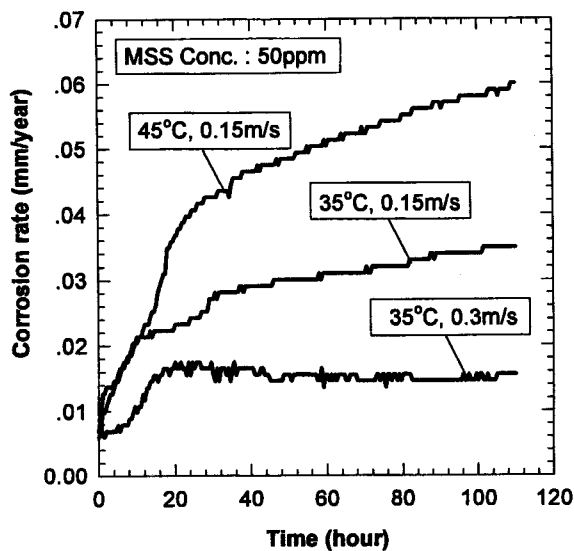


Fig. 11. Effect of temperature and flow velocity on corrosion rate.

Table 2. Weight loss measurement of the carbon steel (STB35) specimens\*

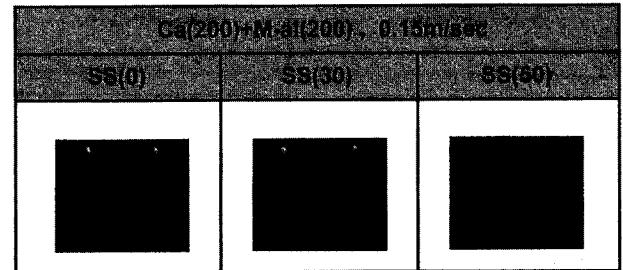
MSS conc. (ppm)	Treatment	Weight <sup>1</sup> (g)		mdd (mg/(0.1 cm) <sup>2</sup> /day)		pH	
		Before	After	Measured	Average	Before	After
0	Prefilming only	11.5399	11.5311	2.8	2.6	8.53	9.11
		11.5879	11.5804	2.4			
30		11.6116	11.5995	3.9	3.9	8.56	9.12
		11.4975	11.4854	3.9			
50	Prefilming + Corrosion inhibitors	11.6205	11.6029	5.6	5.5	8.55	9.12
		11.3756	11.3584	5.4			
0		11.5226	11.5177	1.6	1.5	8.58	8.90
		11.5230	11.5187	1.4			
30		11.4898	11.4843	1.8	1.9	8.56	8.84
		11.5336	11.5274	2.0			
50		11.5776	11.5699	2.5	2.5	8.59	8.88
		11.4090	11.4013	2.5			

\*Experimental conditions were: 35 °C; 0.15 m/sec-equivalent flow velocity; 10-days exposure time

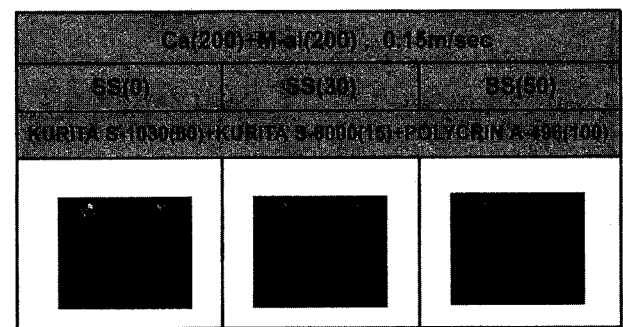
1. Each carbon steel specimen's weight was measured before and after the corrosion experiment. The weight loss was calculated and expressed as mdd in the next column. Each experiment was duplicated.

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biofouling rate, was higher under the low-velocity condition. However, opposite to biofouling it increased at the higher temperature, probably due to the enhanced reactivity of the ionic species at higher temperatures. To simulate the effect of the MSS concentration, a carbon steel STB35 specimen was rotated in a stirred cell filled with cooling water at 35 °C. The rotating speed was adjusted to be equivalent to 0.15 and 0.3 m/sec linear velocity. After 10 days of rotating, the specimen's weight was measured and the degree of corrosion was assessed in MDD (mg per decimeter square



(A)



(B)

Photo 3. Morphology of carbon steel (STB35) test specimens after rotating disk test.

(A) without anti-corrosion treatment, (B) with anti-corrosion treatment

per day). The measurement result is summarized in Table 2, and Photos 3(A) and 3(B) compare the morphology of the untreated and treated specimens. It was confirmed that the higher the MSS concentration, the faster the corrosion proceeded and that the anti-corrosion treatment was effective.

### CONCLUSIONS

For the continuous, on-line measurement of the biofouling phenomenon caused by microbial suspended solids, we have devised and used a biofouling tube made of carbon steel. By measuring the pressure drop increase across the biofouling tube and interpreting the data with the biofilm thickness, we have investigated the effects of the three major parameters on biofilm growth: MSS concentration, temperature, and flow velocity. Of these, flow velocity exhibited the most critical influence, e.g., the biofouling rate at 0.15 m/sec was ca. 4 times higher than that at 0.30 m/sec. Up to 50 ppm, the biofouling rate increased proportionally to the MSS concentration and linearly with time. It was about 1.75 times higher at 35°C than at 45°C, probably because of the diminishing effect of some of the thermolabile microorganisms at 45°C. The biofouling rate expressed in mm/hour ranged from 0.002 to 0.022. The protective prefilming and the anti-corrosion treatment yielded about 60 and 90% reduction in the biofouling rate, respectively. In sum, the biofouling tube apparatus, if installed on-site, could be used to continuously monitor the progress of biofouling and thus to predict timely application of the necessary preventive maintenance programs.

### ACKNOWLEDGMENTS

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