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FOULING AND FOULING MITIGATION ON DIFFERENT HEAT EXCHANGING SURFACES

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ABSTRACT

An apparatus was built to study heat transfer fouling on different heat exchanger pipe surfaces by visually observing the progressive fouling deposition under the same solution conditions. Test pipes removed at the completion of each experiment provided quantitative data on the progressive build-up, time-history of the deposit, as well as the composition of the deposit.

Both hydraulically smooth and artificially roughened test pipes were investigated. These were centrally located in a cylindrical tank with a concentric vertical agitator to give constant and uniform flow conditions near the pipe surface. The calcium sulphate deposition on four different metal surfaces (copper, aluminium, brass, and stainless steel SS 316) where investigated. The results showed that fouling increased with time but at a decreasing rate over set intervals. The deposition also increased with the increasing thermal conductivity of the metal, or the total surface energy. Chemical reaction fouling along with particulate and crystallisation fouling occurred with corrosive chemicals (mixed total fouling) on reactive surfaces, and this was compared with crystallisation-only fouling on nonreactive surfaces.

Bleached Kraft softwood pulp fibres at various concentrations were added to the fouling solution to study their effects on fouling on the hydraulically smooth pipes. Fouling was reduced as fibre concentration increased. Fouling on stainless steel, aluminium, brass, and copper surfaces were all retarded with the low concentration fibre suspensions.

INTRODUCTION

Mineral scale formation on heat exchanger surfaces rapidly increases the thermal resistance and reduces operating service life. It is usually intensified at higher temperatures, because of the inverse temperature-solubility characteristics of some minerals in water. It is a recurring problem in process plants, water heat exchangers, water desalination plants, household equipment, and steam generation units. Five categories of fouling have been proposed depending on the origin of precursors. These categories include biochemical interaction, chemical reaction, particulate, crystallization, and corrosion fouling (Panchal 1999). Five steps in the gradual development of deposits have also been proposed: generation, transportation, attachment, removal, and aging (Epstein 1983). Panchal (Panchal 1999) classified various fouling mechanisms and noted that crystallisation fouling is the deposition of divalent salts by diffusion of ions through the boundary layer to finally form a crystallised deposit on the surface by attraction. Reaction between the cooling fluid and the heat transfer surface causes corrosion fouling.

The present study mainly incorporates crystallisation and particulate fouling, although in some cases corrosion fouling occurred. Hasson (Hasson 1997) reported that asymptotic deposit behaviour observed for mixed salts may be different from solutions of only single salts.

Chemical reactions between the heated metal surfaces and process fluid with chemicals present in it will produce corrosion fouling. Resistance to heat transfer is induced due to in-situ formation of corrosion products. A stable corrosion film can protect the metal surface from high corrosion rates. In interactive fouling there could be a combination of crystallisation with chemical reaction fouling or other mechanisms (Panchel 1999).

Tlili et al. (Tlili, et al., 2008) found that the concentration of calcium sulphate has a stronger effect than temperature in the precipitation process when using a gold layered nickel foil. They used an aqueous calcium sulphate jet impingement method. Demopoulos (Demopoulos, 2009) reported that supersaturation is the important controlling parameter of aqueous precipitation. Mwaba et al. (Mwaba et al., 2006) experimentally investigated calcium sulphate deposition on a flat plate and observed that nucleation started on the downstream side and then propagated upstream. They also observed that the rates of nucleate-front propagation and scale layer growth increased with the surface temperature and decreased with the flow velocity. They have defined super saturation as the ratio of the bulk concentration C_b to the saturation concentration C_s . They

also stated that crystalline calcium sulphate forms on the surface as Gypsum (CaSO₄. $2H_2O$) in the temperature range of 40°C to 98°C. A change of 10 to 20 percent in induction time was observed while experimenting with a copper plate at a Reynolds number of 23,000, initial $C_b = 3.0 \text{ kg/m}^3$ and bulk temperature T_b of 40°C.

Tianqing et al. (Tianqing et al. 1999) observed that both the nucleating and growth rates of $CaCO_3$ particles on a heated surface increases with the concentration of reagent. They also noticed that the diameter of particles increased linearly with time.

Amjad (Amjad 1988) studied gypsum deposition on various metal surfaces and found that the order of the rate of deposition on different metals as; copper > brass > stainless steel.

It is reported that molecules or atoms at the depositing surface interact with those of the solution and the types of forces or interactions depend on the chemistry of both the surface and the fouling liquid (Fletcher 1991). Generally the lower the surface free energy of the depositing surface, the weaker the adhesion of deposit on it (Lindner 1992). Lowenergy surfaces are more resistant to build-up of fouling and easier to clean due to weaker binding at the substratumliquid interface (Baier 1980). The mechanism of fouling deposition reveals that the balance between all possible interactions determine whether a system will foul or not, and fouling will occur when the total interactive force is negative (Zhao and Muller-Steinhagen 1999). Zhao and Muller-Steinhagen (Zhao and Muller-Steinhagen 1999) described surface free energy (surface tension) as a recognised direct measure of intermolecular forces that affect the attraction of the bulk liquid to the surface layer.

Surface roughness leads to a much larger effective contact area than the projected surface area and hence deposit adhesion is stronger (Rankin and Adamson 1973). Surface roughness may also enhance the initial deposition rate by providing sites for crystal nucleation (Taborek, Aoki Yoon and Lund (Yoon and Lund 1994) et al. 1972). reported that no effect of surface roughness was found in the Ra (the arithmetic mean of the departure of the measured roughness profile from the mean line value) range of 0.08 to 0.60µm. McGuire (McGuire 1987) and McGuire and Swartzel (McGuire and Swartzel 1987) showed that surface roughness was not a factor for fouling with their surfaces having Ra values of 0.04, 0.41, 1.93 and 2.31µm for polished SS, rough SS, PTFE coating and alumino-silicate coating respectively. Herz et al. (Herz et al., 2008) also confirmed that calcium sulphate deposition was strongly affected by the degree of surface roughness and the fouling layer on a rough surface was more tenacious than on a smooth surface. They also stated that the higher roughness causes shorter induction periods.

Zhao and Muller-Steinhagen (Zhao and Muller-Steinhagen 1999) reported that mixed $CaSO_4/CaCO_3$ salts foul more heavily than when a single salt exists (CaSO₄ or CaCO₃). They treated metal heat exchanger surfaces by ion implantation, sputtering, electrolytic deposition, or a combination, by sputtering the surface with selected elements. CaSO₄ fouling resistance was reduced by 78 percent.

Junghahn (Junghahn 1964) reported that the free-energy change associated with crystal nuclei formation was much less on a rough surface than on a smooth surface. Other authors (Bott and Gudmundsson 1978, Crittenden and Khater 1987) however, reported higher deposition rates on rougher surfaces.

Gunn (Gunn 1980) investigated 304-stainless steel tubes with different degrees of roughness corresponding to the bonded particles at 80, 120, 180 and 320 Tyler mesh. He maintained the steady-state temperatures of the tube surface and the bulk liquid at 97°C and 95°C respectively. He observed that the rate of nucleation increased with increased surface roughness.

Gill and Nancollas (Gill and Nancollas 1980) studied crystallisation of calcium sulphate on different metal surfaces using a constant heat flux method. They observed more deposition with metals of higher thermal conductivity. They reiterated that the metals with the higher thermal conductivity yielded higher initial steady-state temperatures in the order of Cu > Al > Brass > Stainless steel 304.

Some anti-foulants have been successful in preventing fouling but some have caused undesired product contamination or adverse environmental effects. There are some limited methods to reduce fouling by changing plant operational parameters. Techniques such as electromagnetic, electrostatic and acoustic fields, ultraviolet light, radiation or catalytic treatments have all been tried but with mixed results. The present work is in line with environmentalfriendly procedures by adding solid biodegradable fibre to a fouling liquid to mitigate fouling of metal surfaces.

In the present investigation, the kinetics of calcium sulphate dihydrate (CaSO₄·2H₂O) scale formation on a heat exchanger surface from aqueous solutions have been studied in a reproducible experimental set-up. The effect of bleached Kraft softwood fibre (Kazi, Duffy et al. 2002) on CaSO₄·2H₂O scale formation from metastable supersaturated solutions was also been investigated. It is hoped that the study of the effect of fibre on scale formation on different surfaces with various topographies will show quantitatively the effectiveness of fibre and also throw some light on the mechanisms of inhibition of calcium sulphate dihydrate scale formation in aqueous systems.

The study of fouling on different metal surfaces in a shear field approaches the reality of heat exchangers used in industry. The present work reports the (visual) history of fouling deposition of calcium sulphate with time under the same conditions of heat flux with different types of pipe and surface finishes. It compares crystallisation fouling on stainless steel and aluminium surfaces. Crystallisation and corrosion fouling on copper and brass surfaces are also compared with fouling on stainless steel and aluminium surfaces. Interactive effects of particulate and corrosion fouling are also recorded along with the additional effects of fibre in the fouling solution.

EXPERIMENTAL

(i) The apparatus

The schematic diagram of the experimental apparatus presented in Fig. 1, consists of two flow loops and a stirred tank containing the fouling solution and test pipe. One flow loop comprises a centrifugal pump, rotameter, test section, and a thermostatically controlled water bath to maintain the heating water at a more or less constant temperature ($55\pm1.5^{\circ}$ C). The centrifugal pump (Little Giant Pump Co., USA) having the specification of 2,400 rpm, 0.8 amps, 130 watts motor and of capacity 37 L/minute, with maximum head of 4.5 m. Full details are given in the PhD thesis by Kazi (Kazi, 2001).

The other flow loop is a simple 6mm diameter copper coil for once-through mains cooling water for keeping the bulk temperature at the required value. The temperature drop over the test section was approximately 2.5° C.



Fig. 1 Schematic diagram of the test apparatus.

The fouling solution is contained in the agitated perspex (lucite) cylindrical tank, 202mm diameter and 807mm high. The tank capacity is 17.5L at the top mark. It is graduated from a top mark on the tank; designated A0, (594mm from the bottom). Other level markings from the top A0 are: B0, C0, D0, E0 and F0, which are 90, 162, 234, 342 and 414mm respectively below the top A0 level. The marks were used to locate the fouling sections after different periods of time. The tank level was sequentially dropped to those marks as the experiments progressed.

The stirrer of the tank is an inverted U-shaped PVC impeller with two wings (dimensions 660mm long x 16mm wide x 4mm thick). The speed of the stirrer is controlled with a variable speed drive. The system is isolated from the atmosphere. The bulk fluid temperature was controlled to within $\pm 1^{\circ}$ C.

(ii) Test Specimens

The experimental pipes are 1200mm in length with a wall thickness of 1.2mm and an outer diameter of 12.7mm.

Four metal pipes (stainless steel SS 316, brass, copper, and aluminium) all having the same dimensions were used in this investigation.

The smooth test specimens (Table 1) were used in the as-received condition but before installing in the test rig they were cleaned by rubbing with a water-soaked cloth and flushing with hot water to remove any deposits of grease, oil etc.

Table 1. Hydraulically-smooth and roughened test

Roughened µm Ra
pecimen
SS 316 0.88
Brass 2.30
Copper 2.80
Aluminium 2.90

The roughened test specimens (Table 1) were prepared by sand blasting the surfaces uniformly along the entire length and washed before installing in the test rig. Surface roughness values were obtained using a TAYLOR HOBSON Surfronic 3 instrument. These roughness results are reported as μ m Ra. The instrument was calibrated using a smooth glass surface. The surface roughness of a typical brass roughened test tube was within the limit of variation of 12% along the entire length, indicating the uniformity of the roughened surfaces made in the workshop.

(iii) Data acquisition

nines

A Picolog software program was employed to record the mean tank temperature and the inlet and outlet temperatures of the hot water passing through the test pipe. Both inlet and outlet temperatures of the test pipes were measured using Ktype thermocouples, while the bulk temperature of the solution was measured by using a T-type thermocouple. The program was set to record temperatures every 10 minutes with indication to terminate after 4,000 minutes of data logging.

(iv) Experimental procedure

The tank was filled with distilled water and the hot water flow from the water bath started. The water bath controller was set and the flow rate through the test pipe was also fixed at a predetermined value. The cooling water flow was adjusted to obtain a temperature difference (Δ T) of 15 ± 1°C between the bulk liquid temperature in the tank and the mean surface temperature of the test tube. The average tube surface temperatures of the hot water flowing through the tube. The mixer speed was set to a constant 45rpm so that the surface tangential liquid velocity at the external tube surface was approximately 0.1m/s, roughly equivalent to

previous pipe flow experimental conditions. Heat transfer coefficients were calculated for the various experiments carried out at different tank levels. The calculated heat transfer coefficient for water calibration provided a basis for comparison with values obtained from other experimental conditions, including those with fibre suspensions.

(v) Preparation of calcium sulphate solution

Stoichiometric proportions of calcium nitrate tetrahydrate $Ca(NO_3)_2$ '4H₂O and sodium sulphate Na_2SO_4 were dissolved in distilled water to prepare the aqueous solution of calcium sulphate:

$$Ca(NO_3)_2 H_2O + Na_2SO_4 \rightarrow CaSO_4 H_2O + 2NaNO_3 + 2H_2O$$
(1)

Distilled water was heated to 40°C and then calcium nitrate and sodium sulphate were gradually added sequentially to the solution while continually stirring. The desired concentration of the solution was maintained throughout the experimental runs by taking samples from the sample line at 6 hours intervals. Samples were analysed by titrating with EDTA (Skoog and West 1974) and the tank solution concentration was adjusted accordingly, either by adding more liquid chemicals or by adding more water. When applicable, the fibre suspension concentration was maintained by addition or solution dilution.

(vi) Experiments with water alone and with a 0.01% fibre suspension

An experiment was conducted with water alone to study the heat transfer coefficient of a SS 316 (smooth) test pipe surface for calibration of the test rig. The test was performed at $\Delta T \ 15^{\circ}$ C, U = 0.1 m/s (estimated) and other parameters as described in Table 3. The liquid level in the tank was reduced step-wise and sequentially at a constant interval of 60 minutes from A0 via 1/₂B0, B0, C0, D0, E0, F0, and to the bottom of the tank. Heat transfer data were recorded during each step. Similarly the heat transfer to a 0.01 % fibre suspension in water-only was measured to compare with the results for water using the same sequence.

(vii) Experiments with supersaturated calcium sulphate and fibre suspension

The experimental conditions are summarised in Table 2. The tank level was reduced sequentially so that the test pipes were exposed to the solution for various times. After 25 minutes of running, the level of tank was reduced from A0 to $1/_2$ B0. After 100 minutes the level was lowered from $1/_2$ B0 to B0 and then to C0 after 400 minutes. From A0 to C0 the nucleation and initial deposition could be investigated. From C0 and sequentially via D0, E0, and F0 to the bottom of the tank, the time interval was 1,000 minutes, so that the history of deposition could be visually observed and recorded.

On completion of a run the test pipe was removed, preserved in a container, and air-dried slowly for 7 days.

The deposit patterns, formed deposits, and foreign products could first be examined visually.

The tube surfaces were marked at the prescribed intervals, photographed, and then scraped and the deposit samples weighed. The remaining deposition on each marked length was dissolved in water and concentration was measured by titrating with EDTA (Ethylene-diamine-tetraacetic acid). The dissolved amounts were added to the scraped amount to get the total deposition on each marked interval. The amount in a specified area in each interval was measured and presented in a graphical form.

Table 2. Conditions of the experimental runs.

Parameters	Range of values
U (Outside pipe surface)	0.1 m/s
T _{bulk}	$40 \pm 1^{\circ}C$
ΔT	$15 \pm 1^{\circ}C$
$CaSO_4$ concentration, bulk (C _b)	3.6 g/L
C _s at 55°C	2.16 g/L
Average fibre length	2.53 mm
Fibre coarseness (mass/unit length	0.246 mg/m
RFN Relative fibre number	80
C _{fibre}	0.005 to 0.1 %
pH of solution	7.5

The deposits on the surface were analysed using X-ray diffraction (XRD) to determine the composition of the various deposits. Different crystals have different d spacings. If X-rays of known wavelength λ and measuring angle θ are used (where θ is half of the angle between the diffracted beam and the transmitted beam), the spacing d of the various planes in a crystal can be determined. Therefore by measuring the d spacings, different compounds of deposit could be identified. The XRD patterns are presented by plotting counts (intensity) as a function of 2θ to identify the composition of deposits (Cullity 1959). Bleached Kraft soft wood fibres were used to investigate the effect of fibres on fouling mitigation. Fibres were soaked in water for 24 hours, blended for 20 seconds to disperse them in water, and then added to the fouling solution. Full details are given in the PhD thesis by Kazi (Kazi, 2001).

RESULTS AND DISCUSSION

The experimental procedures are validated in the following sections (i)-(v). The data were also reproduced with good agreement between two similar runs. The results show that the current set-up has yielded similar results to previously-used pipe and annular flow systems.

(i) Water and fibre-water suspension experiments using SS surfaces

The calculated values of the heat transfer coefficients for water as well as a 0.01 mass % suspension of fibre are plotted as a function of time in Fig. 2 for direct comparison.

It can be seen that wood pulp fibres in water reduce the heat transfer coefficient. Similar results for fibre suspensions flowing in pipes have been previously reported (Middis 1994, Muller-Steinhagen et al. 1994, Middis 1994, Kazi, Duffy et al. 1999 and Duffy, Kazi et al. 2000).



Fig. 2 Variation of heat transfer coefficient for water alone, and a 0.01 mass % fibre in water suspension. Bulk temperature 40°C, ΔT 15°C, 45 rpm stirrer (0.1 m/s equivalent).

(ii) Data Reproducibility:



Fig. 3: Fouling deposition on stainless steel SS smooth tubes as a function of time for two different fouling runs at same experimental parameters. Experiments were performed at ΔT 15°C, bulk temperature 40°C and CaSO₄ concentration 3.6g/L.

Fig. 3 represents deposition of $CaSO_4$ on SS smooth surface as a function of time for two different fouling runs under the same experimental conditions (ΔT 15°C, bulk

temperature 40° C, and CaSO₄ concentration 3.6g/L). It is observed that data reproducibility are good and remain within the confidence level of 95 percent with an average rms error less than 5 percent. The little variation is due to incursion of foreign particles in the solution during the course of experiment as reported by Gill and Nancollas (Gill and Nancollas, 1980).

(iii) Fouling in CaSO₄ solution - solution concentration effect (without fibre)

The deposition of calcium sulphate from two different supersaturated solution concentrations on the stainless steel SS 316 (smooth) surface as a function of time was studied and the results are presented in Fig. 4.



Fig. 4 Fouling deposition as a function of time for two different concentrations of $CaSO_4$ solution. ΔT 15°C and bulk temperature of 40°C.

It is observed that deposition increases with the increasing solution concentration as well as the time of exposure of the surface to the solution. This is caused by the increased amount of crystallisation in the boundary layer and increased total attractive force to the surface. Again similar results were obtained by Kazi et al. (Kazi, Duffy et al. 1998), Middis (Middis 1994) and Bansal (Bansal 1994).

(iv) Fouling in CaSO₄ solution in the presence of fibre (fibre concentration effect)

The effect of varying the fibre concentration in the CaSO₄ solution (3.6g/L) was investigated for up to 4,000 minutes exposure using the SS 316 pipe surface. The results are presented in Fig. 5.

Fibre concentration was varied from 0 to 0.1% in three steps. The results show similar trends of deposition. The slopes of the curves decrease with increasing fibre concentration. By adding 0.005%, 0.01% and 0.1% fibre separately to the supersaturated CaSO₄ solution, the fouling depositions were reduced 24.3%, 78.4% and 99.7% respectively at the specified final time of observation of 4,000 minutes.

The results are in concert with those obtained in pipe flow studies previously (Middis, Paul et al. 1997, Kazi, Duffy et al. 1998). Fibres reduce the rate of deposition by 'shielding' the surface from approaching crystals formed in the boundary layer, by sustained micro-mixing, and by the continuous physical collision with the heated surface. Fibres affect the turbulent eddies which reduce the rate of mass transfer of the fouling elements to the heated wall (Kazi, Duffy et al. 1998).



Fig. 5 Fouling deposition as a function of time for different fibre concentrations on a SS 316 (smooth) surface, ΔT 15°C, solution conc. 3.6g/L, bulk temperature 40°C.

(v) Fouling with CaSO₄ on various smooth metal surfaces (without fibre)

The influence of the nature of the metal (copper, aluminium, brass and stainless steel) on the deposition of calcium sulphate scale was studied in a series of experiments.

Fig. 6 shows the calcium sulphate deposition on four different heat exchanger surfaces as a function of time. The deposition on different metal substrates was consistent with their thermal conductivity values, i. e. copper > aluminium > brass > stainless steel.

It is also observed that the deposition tends to be asymptotic which is consistent with pipe flow investigations (Kazi, Duffy et al. 2002). Forster and Bohnet (Forster and Bohnet 1999) found that the deposition on different metal substrates were consistent with their values of the polar component of the surface free energy, i.e. copper > aluminium > SS > brass. Amjad (Amjad 1988) obtained similar results for calcium sulphate deposition on copper, brass and stainless steel surfaces at a higher solution concentration of 4g/L. It was also observed that the fouling on metal surfaces is asymptotic. These experimental results also appear to correlate well with thermal conductivity, with deposition decreasing as thermal conductivity decreases.



Fig. 6 Deposition of $CaSO_4 2H_2O$ as a function of time and metallurgy of heat exchanger. ΔT 15°C, bulk temperature 40°C, solution conc. 3.6g/L and

(vi) Fouling with CaSO₄ on roughened metal surfaces (without fibre)

Data for the deposition of $CaSO_4$ on smooth and roughened metal surfaces are presented in Fig. 7. Clearly it is observed that fouling deposition increases with increasing roughness.



Fig. 7 Fouling deposition as a function of time for Cu, Al, brass and SS test pipes both smooth (S) and sand blasted (SB). ΔT 15°C, bulk temperature 40°C, solution conc. 3.6g/L and 0.1 m/s.

The order of the ranking of deposition on the roughened surfaces is consistent with the order of deposition recorded on smooth surfaces of those metals, and the trends in the fouling data are also asymptotic. The results are in agreement with the observation of Junghahn (Junghahn 1964) with respect to the free energy change associated with crystal nuclei formation which is much less on a rough surface than on a smooth surface.

Present observations also qualify the inference of Junghahn that edges and sharp points are centres of high surface energy causing preferential nucleation sites. The rough surfaces produce greater deposition than the smooth surfaces due to protected zones in the cavities or pits where flow velocities are very low and nucleation rates are higher (Rankin and Adamson 1973).

Once crystallisation has commenced, the higher adhesive forces between the fouling layer and the surface influence the removal rate and lead to higher asymptotic fouling values for the rough surfaces.

(vii) Fouling with CaSO₄ on smooth surfaces in the presence of fibre



Fig. 8 Fouling deposition as a function of time on Cu, brass, and SS 316 smooth surfaces exposed to solution with-and-without fibre in suspension. ΔT 15°C, bulk temperature 40°C, 0.01% fibre, solution conc. 3.6g/L, 0.1m/s.

Three different smooth test specimens (copper, brass, SS 316) were investigated in an aqueous $CaSO_4$ solution of concentration 3.6g/L with 0.01% fibre in suspension. Fig. 8 shows the deposition of $CaSO_4$ as a function of time. The deposition increases with increasing thermal conductivity of the material i.e. Cu shows the highest deposition followed by brass, and then SS 316.

Fibres in turbulent motion away from the boundary layer affect the boundary layer which reduces the rate of mass transfer of the foulant to the heated tube surface (Kazi, Duffy et al. 2002). It is observed that the addition of even a very small amount of fibre in the aqueous solution of $CaSO_4$ consistently reduces the foulant deposition on different metal surfaces at any specified time interval.

(viii) Effect of fouling liquid on surface topography

Table 3 summarises the effect of corrosion on different smooth metal surfaces. No corrosion effects on SS and Al

surfaces were observed. Brass and Cu surfaces experienced corrosion and that generated change at the surfaces which caused both crystallisation and corrosion fouling. Although there were no measurable roughness changes in Al and SS smooth surfaces, the results could be compared with total-fouling deposition on smooth surfaces. The achieved roughness changes all lie within the tolerable limit (Yoon and Lund 1994, McGuire 1987) of surface alterations up to a comparable time of exposure to fouling liquid.

Table 3. Surface roughness change of smooth test specimens.

Time (min)	SS 316	Aluminium	Brass	Copper
0 000	0.25	0.22	0.48	0.25
1 000	0.25	0.22	0.64	0.46
2 000	0.25	0.22	0.98	0.95
3 000	0.25	0.22	1.38	2.38
4 000	0.25	0.22	1.38	2.38

Similarly, surface roughness changes of roughened metal surfaces are also due to corrosion as presented in Table 4. The roughened surfaces of selected materials Cu, Al, brass, and SS 316 follow similar trend as discussed earlier in the case of smooth surfaces.

Table 4. Surface roughness change of rough test specimens.

Time (min)	SS 316	Aluminium	Brass	Copper
0.000	0.88	2 90	2 30	2.80
1 000	0.88	3 20	2.50	2.80
2 000	0.88	3.80	2.72	3.48
3 000	0.88	3.80	2.95	4.07
4 000	0.88	3.82	2.96	4.08

(ix) Visualisation of the surface and crystal morphology

The photographs in Fig. 9 show the white deposits formed on stainless steel SS 316 and aluminium surfaces, indicating only crystallization fouling. The deposits formed on the brass surface however have green stains along with white heavy encrustations of calcium sulphate, showing some surface chemical attack and hence some chemical reaction fouling.

The green crystals are $CuSO_4$ formed by reaction between the Cu component in brass and $CaSO_4$. Similarly the deposition on the copper pipe surface also has a green layer along with the white deposition showing combined effect of crystallization and chemical reaction fouling. Thus in industry, it could be inferred that the cooling water affect on corrosion-prone heat exchanger surfaces are the combined total effect of fouling.

In these experiments fibre is used to mitigate fouling deposition on metal surfaces. So when fibre is used, the depositions on SS and aluminium surfaces were of similar nature.

Fibre is mixed with deposits and forms a composite. On the brass surface, $CuSO_4$ and $CaSO_4$ formed composites with fibre, but on the copper surface reddish and black stains of CuO and Cu₂O also formed in the composites. However the total deposition order on different metal surfaces does not change due to chemical reactions.

Fig. 9(1–4) Deposition on smooth SS 316 (1), aluminium (2), brass (3) and copper (4) surfaces. ΔT 15°C, bulk temperature 40°C, solution conc. 3.6 g/L, 4,000 min.

Fig. 9(1) and Figs. 10(a) to 10(c) represent the crystalline deposition on the SS 316 (smooth) surface after 4,000 minutes of exposure to 3.6 g/L aqueous calcium sulphate with fibre suspensions of 0%, 0.005%, 0.01% and 0.1% concentration respectively.

There is absolutely no significant deposition on the stainless steel tube surface with 0.1% fibre in fouling liquid. It is also observed that fibre consistently mitigates fouling on metal surfaces (Refer to Fig. 5).

Fig. 10(d) and 10(e) represents the deposition on brass and copper (smooth) surfaces after 4,000 minutes of exposure to 3.6g/L aqueous CaSO₄ with fibre suspension of 0.01%. It is observed that the order of total deposition on SS 316 Fig. 10(b), brass Fig. 10(d), and copper Fig. 10(e), all for 0.01% fibre present, remains the same as the case where no fibres are present.

Deposition on SS 316 surface is $CaSO_4.2H_2O$ but on brass surface it is accompanied with a green layer of $CuSO_4.2H_2O$ formed due to the reaction of Cu component of brass with $CaSO_4$ solution.

In the case of the copper surface, $CaSO_4.2H_2O$ is formed on the surface accompanied with the green stain of $CuSO_4.2H_2O$. In addition to the green stain, reddish and black stains of CuO and Cu_2O are also obtained on the copper surface due to reaction of Cu with $CaSO_4$ solution.







Fig. 10(b)

Fig. 10(c)

Fig. 10(a–c) Crystal growth on SS 316 (smooth) surface exposed to solution of CaSO₄, conc. 3.6g/L, Δ T 15°C, bulk temperature 40°C, 4,000 minutes exposure and 0.005%, 0.01% and 0.1% fibre respectively.



Fig. 10(d)

Fig. 10(e)

Fig. 10(d-e) Deposition on brass and copper (smooth) surface exposed to solution of CaSO₄, conc. 3.6g/L, ΔT 15°C, bulk temperature 40°C, 0.01% fibre and 4,000 minutes exposure.

Fig. 11 presents XRD spectra of the deposits of copper and brass after 4,000 minutes exposure. The deposition samples include those from smooth and sand-blasted roughened copper pipes with no fibre in the fouling solution (see Fig. 7), as well as deposits from the smooth copper and brass pipes with 0.01 percent fibre in suspension (see Fig. 8). The results show that the deposits on the hydraulically smooth copper pipe with-and-without fibre in solution are comprised of hydrated calcium and copper sulphates and cuprous oxide Cu_2O . Visually there are black patches on the surface.



Fig. 11 Counts as a function of 2 Theta of XRD for crystal and fibre crystal composite samples deposited on different (Cu and brass) metal surfaces. The experiments were conducted at ΔT 15°C, bulk temperature 40°C, CaSO₄ concentration of 3.6g/L and fibre concentration of 0 to 0.01 percent.

The deposit from the roughened, sand-blasted copper surface is a composite of $CaSO_4.2H_2O$ and $CuSO_4.2H_2O$. Visual inspection of the surface shows occasional reddish stains along with black patches but the XRD could not identify exactly if it were cupric oxide CuO, as only a minute amount was present.

XRD analysis of the composite deposition on brass smooth surface after fouling run with 0.01 percent fibre in suspension (see Fig. 8) shows hydrates of $CaSO_4$ and $CuSO_4$. Clearly the nature of the composite deposits varies with different metal surfaces but the fouling resistance is the cumulative effect of deposition on the surface.

The surface chemical reaction also affected the wall thickness and surface roughness, and hence the fouling rate was also changed. After experiments of 4,000 minutes duration, the roughness of the initially smooth brass and copper surfaces were both increased from 0.48 to 1.38 μ m Ra, and from 0.25 to 2.38 μ m Ra respectively, but the changes were not very high or far beyond acceptable limits (Table 3) (Yoon and Lund 1994, McGuire 1987, McGuire and Swartzel 1987 and Mwaba et al., 2006). There were no measurable roughness changes for the aluminium and SS surfaces.

The crystal size of deposits on different metal surfaces was studied under the microscope. Fig. 12(a-d) represents the comparison of crystal size by microscopic view of the crystals formed on different metal (smooth) surfaces (SS 316, brass, aluminium, and copper) after 1000 minutes exposure to a solution of CaSO₄ having a concentration of 3.6 g/l. It is found that crystals formed on copper, aluminium, brass, and SS surfaces increase in average magnitude in the same order of thermal conductivity: copper > aluminium > brass > SS 316 surfaces. Similar results were reported by Liu et al. (Liu, T., Shi, Y. and Wang, X., 1999) for CaCO₃ deposition on heated surfaces. They stated that bigger crystals could be found on surfaces where rate of deposition is higher.





Fig. 12(a) SS 316.

Fig. 12(b) aluminium.



Fig. 12(c) brass.



Fig. 12(d) copper.

Fig. 12(a-d) Microscopic view of crystals formed on SS 316, brass, aluminium and copper smooth surfaces. 1,000 minutes exposure to solution of CaSO₄, solution conc. 3.6g/L, $\Delta T 15^{\circ}C$ and bulk temperature 40°C.

The crystal sizes of deposits on the brass surface have been systematically studied. It is reported (see Fig. 6) that the total fouling mass per unit surface area increased with time in the specified cases in the range of investigation.

Figs. 13 and 12(c) represents the gradual development and growth of crystal size with time on a smooth brass test piece. The largest sized crystals are obtained at 4,000 minutes of exposure of the tube specimen to the fouling liquid. It is also observed that at 1,000 – 4,000 minutes the deposit contains many crystals closely packed together, and large crystals are surrounded by many smaller randomlyoriented crystals. Similar results were obtained by Tianqing et al. (Tianqing, Yuwen et al. 1999) for the deposition of $CaCO_3$. It can be noted that the photos in Fig. 13 also indicate the crystal morphology.



25 minutes

100 minutes



400 minutes







4,0

4,000 minutes

Fig. 13 Growth of crystal size with time (minutes) on a smooth brass test piece. Experiments performed at CaSO₄ concentration 3.6g/L, Δ T 15°C and bulk temperature 40°C.

However, in this case the surface attack is not as marked as in the case of the deposit on a rough brass surface. Thus for short term experimental purposes, the total deposition on brass and copper are cumulative effect of crystallization and surface reaction fouling. Extra care should be given in industry during long term operation if reaction fouling occurs because tube corrosion will be ongoing.

CONCLUSIONS

It can be concluded that:

- 1. The extent of fouling on different metal surfaces increases with increasing thermal conductivity of the metal (copper > aluminium > brass > stainless steel).
- 2. Increasing surface roughness increases fouling.
- Fibres in solution retard fouling on smooth surfaces. The induction period is also increased as previously observed in separate pipe flow and annular flow experimental systems.
- 4. The effect of fibres on fouling mitigation is consistent, and irrespective of the tube material used.
- 5. Surface characteristics are altered by corrosive attack and this promotes fouling and reduces the effectiveness of mitigation. Thus when different metal surfaces are compared, a single category of fouling could not be specified.
- 6. Movement of insoluble corrosion products and their detachment and re deposition need to be studied to generate realistic models for industrial use. The effect of soluble corrosion products on the generation of fouling precursors also needs to be studied.

NOMENCLATURE

- C_{fibre} Fibre concentration in percent
- Ra The arithmetic mean of the departure of the measured roughness profile from the mean line value
- T_{bulk} Solution bulk temperature in °C
- ΔT Temperature difference in °C
- U Velocity in m/s
- C_b Bulk concentration of CaSO₄
- C_s Saturation concentration of CaSO₄ at surface temperature
- RFN Relative number of fibres per unit mass

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