

THE INVESTIGATION OF PARTICULATE CORROSION PRODUCT DEPOSITION ON HEAT TRANSFER SURFACES: A COMPARISON OF EXPERIMENTS AND THEORY AND A PRELIMINARY STUDY OF THE REMOVAL MECHANISM

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ABSTRACT

The research program at the University of New Brunswick (UNB) into the fouling of heat exchangers in cooling water systems began in the early 1990s. Since then, experiments and numerical simulations (using the fluid dynamics codes PHOENICS and FLUENT 4.5) have investigated the deposition of oxide particles suspended in water at atmospheric pressure onto heated surfaces. This paper summarizes the program and tests the mathematical model that was proposed in 2007 against the experimental results. The experiments were carried out under isothermal conditions and various modes of heat transfer: non-boiling, subcooled boiling and bulk boiling. Sub-micrometre particles of iron oxides (magnetite and nickel ferrite) were synthesized and added to a recirculating loop, where their deposition from suspension along an Alloy-800 tube at a heat flux up to 240 kW/m² was studied. The mathematical model considers deposition and concomitant removal of particles and consolidation of deposits as a comprehensive deposition mechanism under transport-controlled conditions. It indicates that size, nucleation frequency and nucleation site geometry of bubbles play an important rôle in the global deposition flux. Predictions of the model are in general agreement with the experimental results. Recently, the mechanisms have been tested experimentally under conditions favouring attachment control rather than transport control. Similar results were obtained for deposition but not for removal.

INTRODUCTION

Fouling is known as the formation of process-related deposits on heat transfer surfaces. It has been recognized as a universal problem in design and operation that leads to an increase in the resistance to heat transfer and reduces the effectiveness of heat transfer equipment. In addition, the deposition of corrosion products can lead to the concentration of impurities, resulting in localized corrosion which can damage components.

The mechanisms of particulate fouling have been described in previous studies (Kern and Seaton, 1959; Gudmundsson, 1981; Epstein, 1988). Most authors based their fouling mechanisms on the original Kern-Seaton approach, which described fouling as a competition between the rates of deposit growth and removal (Lahey, 1992).

Previous studies in our laboratories on the fouling of heat exchangers by iron oxides worked toward a comprehensive mechanism of the deposition via four steps, namely: transport, attachment, removal, and consolidation (Basset et al., 2000; Cossaboom and Lister, 2005; Lister and Cussac, 2009). Key experiments in the series have involved radiotracing methods. Because of their complexity, replicate experiments have not been attempted. The goal of this paper is to summarize that previous work and apply the particulate fouling model (Lister and Cussac, 2009) that was proposed in 2007 to the experimental results (Basset et al., 2000; Callamand, 1999; Capentier, 2002; Cossaboom and Lister, 2005; McCrea, 2001). In particular, the removal and consolidation mechanisms of nickel ferrite particles on Alloy-800 heat transfer surfaces in water under subcooled boiling conditions are addressed. It is noted that previous experiments considered the simultaneous deposition and removal of particles; i.e., the behaviour of deposits when a continuous source of suspended material was available in the coolant. In order to shed more light on the mechanisms, particularly consolidation, when particle attachment rather than transport is controlling, a preliminary study of deposit behaviour when suspended particles were removed from the coolant was undertaken.

PARTICULATE FOULING MODEL

Kern and Seaton (1959) first formulated a mathematical model for particulate fouling. The net fouling rate is the difference between the rates of deposition and removal of particles from the heat transfer surface:

$$\frac{dm_f}{dt} = \Phi_d - \Phi_r \quad (1)$$

where m_f is the mass of the deposit in kg/m² and Φ_d and Φ_r are the deposition flux and the particle removal flux in kg/m²s, respectively.

When a linear dependence with the concentration in the bulk liquid is assumed (Epstein, 1988):

$$\Phi_d = K_d C_b \quad (2)$$

where K_d is the deposition velocity (m/s) and C_b is the concentration of depositing material in the bulk liquid (kg/m³).

Lister and Cussac (2009) proposed a model of iron oxide deposition, based on experimental results from the water-cooled fouling loop at UNB. The general equation for isothermal, non-boiling and subcooled boiling conditions is:

$$m(t) = \frac{\Phi_d}{k_r} (1 - e^{-k_r t}) \quad (3)$$

where k_r is the removal constant (s^{-1}).

The consolidation term is added when the time is greater than the critical time, t_c , which is the time when consolidation first occurs.

The amount of iron oxide deposited under bulk boiling can be obtained from the modified Turner and Klimas model (2001) as follows:

$$m(t) = \frac{\Phi_d}{k_r + k_c} \left[k_c t + \frac{k_r}{k_r + k_c} (1 - e^{-(k_r + k_c)t}) \right] \quad (4)$$

where k_c is the consolidation constant (s^{-1}). The quantities Φ_d , k_r and k_c are calculated from various correlations under different modes of heat transfer as described below.

The diffusion from the deposit to the bulk is a removal mechanism that becomes unimportant under boiling conditions but is dominant for non-boiling heat transfer systems (Lister and Cussac, 2009). The diffusion coefficient, K_{diff} , is based on the Levich model (1962) with an empirical boiling parameter BR defined by McCrea (2001):

$$K_{diff} = (1 + BR) \times 0.8 \frac{u^*}{Sc^{3/4}} \quad (5)$$

where u^* is the friction velocity equivalent to $u\sqrt{f}/2$, u is the velocity of the fluid, f is the friction factor obtained from the classical Blasius equation ($f = 0.791/Re^{0.25}$) and Sc is the Schmidt number. BR takes into account the enhanced deposition due to boiling as defined by:

$$BR = \frac{0.05q}{R_b L} \frac{(T_{film} - T_{sat})}{T_{sat}} \quad (6)$$

where q is the heat flux (kW/m^2), L is the latent heat of vaporisation (J/kg), T_{film} and T_{sat} are the film and saturation temperatures and R_b is the bubble radius computed from microlayer evaporation modelled by Asakura (1978).

ISOTHERMAL AND NON-BOILING

The thermal hydraulics of the flow system for the fouling loop were studied by Margotin (1994). He determined the flow properties of the test section (a vertical glass column 1.5 m long and 9.93 cm diameter containing a 25 cm length of Alloy-800 tubing, 1.27 cm in diameter fitted with an electrical heater) using PHOENICS, a Computational Fluid Dynamics (CFD) code. The accuracy of the model was assessed by measuring velocities of suspended particles of magnetite with a laser-Doppler anemometer. It was assumed that the discrete phase had a negligible effect on the flow properties. The study involved a Lagrangian calculation to predict the magnetite behaviour under isothermal condition. Two sizes of particle (0.5 and 1.0 μm diameter) were used in the experiments. The results

showed that the smaller the particle, the greater the probability of deposition.

Deposits formed under isothermal and under sustained heat transfer but non-boiling conditions are similar and are associated with surface roughness. For colloidal particles, diffusion is the dominant transport mechanism. In this regime, the particles move with the fluid and are carried to the surface by Brownian motion. For systems of high Schmidt number, the transport coefficient under diffusion-dominated conditions was derived by Callamand (1999). Using a modified Cleaver and Yates (1975) model, he computed transport coefficients that were in good agreement with the experimental results. Without heat flux, under constant temperature, BR is zero and K_d is K_{diff} . The removal in this case is the diffusion from the deposit to the bulk, which can be obtained from:

$$k_r = 0.8 \frac{u^*}{Sc^{3/4}} \quad (7)$$

Magnetite deposition on Alloy-800 heat transfer surfaces was further studied under non-boiling conditions by Basset et al. (2000). Sub-micrometre magnetite particles suspended in solution at pH_{25°C} 7.5 deposited under nominal transport control by diffusion.

Thermophoresis is the thermal force that moves fine particles down a temperature gradient so that cold walls attract and hot walls repel colloids. The thermophoretic force increases with the temperature gradient but decreases with increasing particle size. Thermophoresis has been shown to be an effective mechanism for particle removal when the size of the particles is less than 2 μm . It is given by McNab and Meisen (1973):

$$K_{th} = 0.26 \frac{1}{2\lambda_w + \lambda_p} \frac{v_w}{T_{sat}} q \quad (8)$$

where q is the heat flux, λ_w and λ_p are thermal conductivities of water and the particle and v_w is the kinematic viscosity of the water. The deposition rate includes a corrective term to take thermophoresis into account. K_d then becomes:

$$K_d = K_{diff} - K_{th} \quad (9)$$

BOILING

Particulate fouling is generally more severe under boiling conditions than under convective heat transfer. The deposition of iron oxide during subcooled boiling was studied by Basset et al. (2000). Experiments into magnetite deposition were conducted with a water temperature of 90°C and a flow rate of 12 L/min in a recirculating water loop similar to that shown schematically in Figure 5. The magnetite concentration was kept at 0.005 kg/m^3 . Under various pH conditions, the deposition rate reached its maximum at a pH of 7.5-8.5, suggesting that the deposition is transport-controlled in this region. Basset et al. postulated that release occurs concomitantly with the movement of particles to the surface via the vapour-liquid interface of growing bubbles.

McCrea (2001) showed how deposition in bulk boiling modifies the heat transfer surface. The highest deposition

rate of magnetite in her experiments was found at a pH of 5-6. The evolution of the deposition with time was also studied. The deposition was linear during the first 50 hours for pH between 3.5 and 11.0.

Carpentier (2002) simulated the thermal hydraulic conditions of the fouling loop using the CFD code FLUENT 4.5. A two-fluid model associated with dispersed turbulence was used for the prediction of flow under subcooled boiling. In total, the formation and behaviour of 2,500 bubbles were tracked. Average bubble diameter decreased with increasing heat flux and the deposition flux of magnetite under subcooled boiling was found to vary with the fraction of vapour in the test section. Possibly because no removal step was incorporated in the model, deposition was over-predicted by a factor of ten.

Particle removal from the surface occurs differently under different boiling conditions. Cossaboom and Lister (2005) observed that particle removal is more pronounced for the deposition of magnetite under sub-cooled boiling (as found by Basset et al. (2000)) than the deposition of nickel ferrite under bulk boiling. Lister and Cussac (2009) suggested that bubble collapse influences the deposit by creating a certain amount of turbulence in the viscous sub-layer and disturbs the flow in the deposit vicinity. Under sub-cooled boiling conditions, the bubbles collapse close to the surface and the outer layers of deposit are permanently maintained in a labile and removable state. Thus, deposit formation under boiling conditions involves the turbulence controlled by bubble formation and detachment:

Removal mechanism during boiling

Factors in the removal step include dissolution, erosion and spalling. Under flow-boiling condition, particle removal involves the action of hydrodynamic forces and thermal stresses.

Epstein (1997) reported that viscous shear is mainly responsible for the removal of deposits during particulate fouling. However, the re-entrainment of particles was found to be insignificant within the experiment range $0.5 < d_p^+ < 1.3$ (Yung et al., 1989), where d_p^+ is the dimensionless particle diameter ($d_p^+ = d_p u^*/\nu$), d_p is the average particle diameter, u^* is the friction velocity and ν is the fluid kinematic viscosity.

Under boiling conditions, the rate of accumulation of particles on the surface is seen to decrease with time initially and it is postulated that some of the deposit remains available for removal as an increasing fraction becomes consolidated (Basset et al., 2000; Cossaboom and Lister, 2005). The fraction consolidated is different between subcooled and bulk boiling. The experiments in the current study (described later) look at the removal of colloidal nickel ferrite particles under subcooled boiling. "Non-concomitant particle removal" occurs when a deposit is exposed to a particle-free fluid flow. It is required in this study as an input to the deposition mechanism for nickel ferrite particles, which was formulated from radiotracing experiments involving release with concomitant deposition (Lister and Cussac, 2009). The experimental results are used to determine the non-concomitant removal constant under particle-free fluid flow, k_{r0} (in units of s^{-1}).

Consolidation mechanism during boiling

A previous study on magnetite particles deposited during subcooled boiling (Basset et al., 2000) led to the conclusion that the entire deposit was labile and could be removed/exchanged and that consolidation did not occur. However, Lister and Cussac (2009) suggested that consolidation also takes place in this boiling regime but begins only after a certain time and then only a fraction of the deposited particles is consolidated. Their model takes into account the portion of the heat transfer surface that is covered by active nucleation sites and describes the evaporation rate of bubbles as a function of the boiling intensity. By contrast, for bulk boiling, Cossaboom and Lister (2005) had suggested that the consolidation starts at the beginning of the deposition.

A consolidation parameter is related to the labile portion of the deposit by (Cussac and Lister, 2009):

$$m_{\text{consolidation}}(t) = m_{\text{labile}} \times 10^{-4} \left(\frac{t}{t_c} - 1 \right) \quad (10)$$

where m_{labile} is a function of surface area of a nucleation site, active nucleation site density, heat flux and latent heat of vaporisation; t_c is the critical time when consolidation first occurs.

The calculations of the rate constants for removal and consolidation (k_r and k_c) under both boiling regimes can be found in detail in Lister and Cussac (2009).

PARTICULATE FOULING MODEL VALIDATION

The particulate fouling model (Lister and Cussac, 2009) predicts the amount of iron oxide deposited on Alloy-800 heat transfer surfaces. It had been applied to the radiotracing results of Basset et al. (2000) and Cossaboom and Lister (2005) and fitted them very well. Operating conditions from other experiments undertaken in the fouling loop are shown in Table 1 and will be applied to the model.

Table 1. Summary of the experimental operating conditions for the model evaluation.

Run #	q	T _b	C _b	d _p × 10 ⁶	Flow Rate	pH	Time
Isothermal:							
1	0	90	0.005	0.6	12	7.5	60
Non-boiling:							
2	100	90	0.005	0.6	12	7.5	50
Subcooled boiling:							
3	100	96	0.005	0.6	12	7.5	50
Bulk boiling:							
4	240	96	0.005	0.6	12.42	7.0-8.0	50
5	240	96	0.005	0.8	12.40	~7.0	25

Note: q is in kW/m², T_b is in °C, C_b is in kg/m³, d_p is in m, Flow Rate is in L/min and Time is in h.

All experiments were done using magnetite particles except Run #5, which used nickel ferrite particles. Note that at the pHs of the experiments the deposition is expected to be under transport control.

Isothermal and non-boiling

The experimental results from Run #1 (isothermal case) and Run #2 (non-boiling heat transfer case) are presented in Figure 1 and Figure 2, respectively, along with predictions of the particulate fouling model.

For the first 50 hours in both runs, the measurements indicate an almost constant deposition velocity, K_d (defined as the deposition rate Φ_d at time zero divided by the bulk concentration, C_b), which amounts to $0.22 \mu\text{m/s}$ for Run #1 and $0.32 \mu\text{m/s}$ for Run #2.

The deposition behavior can be explained using a diffusion mechanism and accounting for thermophoresis. K_d calculated from Equation (5) is $0.927 \mu\text{m/s}$ and $0.554 \mu\text{m/s}$ in Figures 1 and 2, respectively. In the isothermal case, Figure 1, the K_d (which is equivalent to K_{diff} calculated from Equation (5)) is four times higher than the results. The particulate fouling model in Equation (3) fits the experimental data better when modified with the Cleaver and Yates correlation (Cleaver and Yates, 1975) for systems of high Schmidt number. The transport coefficient for diffusion-dominated transport is then:

$$K_d = K_{\text{diff}} = 0.084 \frac{u^*}{Sc^{2/3}} \quad (11)$$

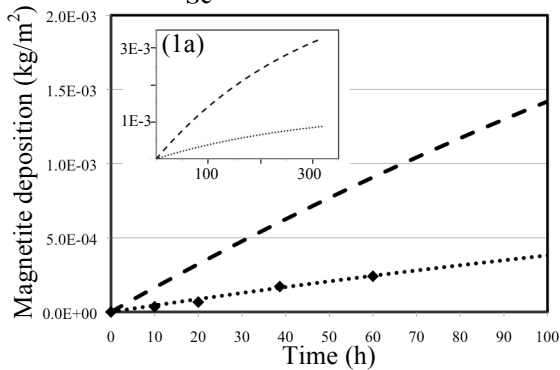


Fig. 1 Evolution of the deposit mass with time under isothermal conditions, (1a) shows long-term deposition under the same operating conditions (--- Particulate fouling model, ... Particulate fouling model with the Cleaver and Yates modification, ♦ Run #1).

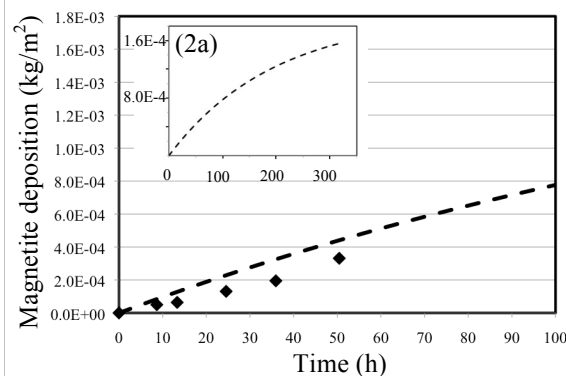


Fig. 2 Evolution of the deposit mass with time under non-boiling conditions with $q = 100 \text{ kW/m}^2$, (2a) shows long-term deposition under the same operating conditions (--- Particulate fouling model, ♦ Run #2).

After the modification, K_d becomes $0.249 \mu\text{m/s}$, which

is in good agreement with the experiments of Basset et al. (2000) in Figure 1. That there is a thinner deposit under constant heat flux than under isothermal conditions can be explained via a diffusion mechanism while accounting for thermophoresis effects. Thus, K_{th} and K_{diff} calculated from Equations (8) and (11) are $0.866 \mu\text{m/s}$ and $1.416 \mu\text{m/s}$, respectively. The agreement with experiment is fair (see Figure 2).

Subcooled boiling

Under subcooled boiling conditions, the mechanisms of deposition are controlled by microlayer evaporation, bubble nucleation and bubble growth. Figure 3 presents data from Basset et al. (2000) for magnetite under subcooled boiling conditions. The present model is based on nucleation site stifling and site-reactivation. When a site stifles or stops generating bubbles, a new site is activated in order to maintain constant site density. Values obtained from the particulate fouling model under subcooled boiling conditions for K_d , k_r and k_c are $11.5 \mu\text{m/s}$, $2.70 \times 10^{-6} \text{ s}^{-1}$ and $9.03 \times 10^{-7} \text{ s}^{-1}$, respectively. The predictions fit the data reasonably well (Figure 3).

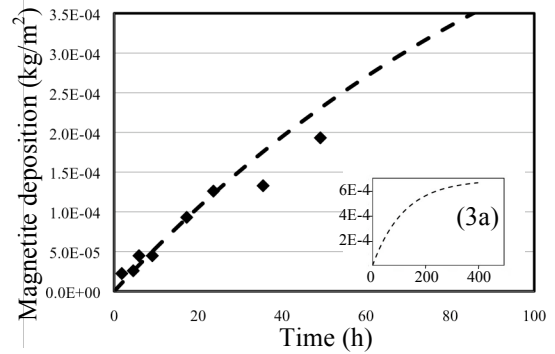


Fig. 3 Evolution of the deposit mass with time under subcooled boiling conditions with $q = 100 \text{ kW/m}^2$, (3a) shows long-term deposition under the same operating conditions (--- Particulate fouling model, ♦ Run #3).

Bulk boiling

McCrea (2001) studied the deposition of magnetite particles onto Alloy-800 under bulk boiling conditions (Run #4) and Cossaboom and Lister (2005) studied nickel ferrite deposition under similar conditions in Run #5. Results from Cossaboom and Lister are somewhat lower than McCrea's (see Figure 4).

Values obtained from the particulate fouling model under bulk boiling conditions for K_d , k_r and k_c are $5.28 \mu\text{m/s}$, $2.88 \times 10^{-5} \text{ s}^{-1}$ and $6.86 \times 10^{-6} \text{ s}^{-1}$, respectively. The comparison with the data indicates an over-prediction.

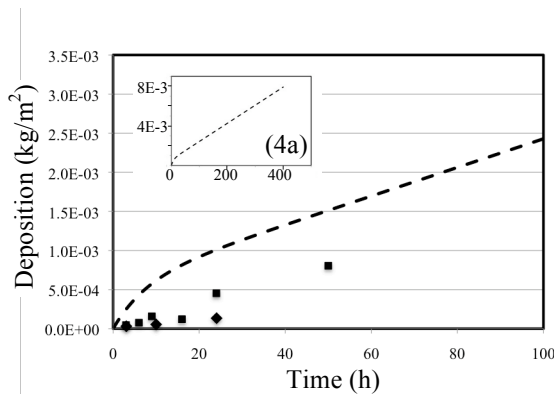


Fig. 4 Evolution of the deposit mass with time under bulk boiling conditions with $q = 240 \text{ kW/m}^2$, (4a) shows long-term deposition under the same operating conditions (--- Particulate fouling model, ■ Run #4, ♦ Run #5).

EXPERIMENTAL TECHNIQUES

Nickel Ferrite Solid-State Synthesis

In order to obtain data on the non-concomitant removal of particulate deposits (when the concentration of particles in suspension or “source term” is removed from the fluid) a further set of experiments was performed. Well-defined nickel ferrite particles, uniform in size and shape and resembling corrosion products found in nuclear reactor systems (Lister and Venkateswaran, 1995) were synthesized by a solid-state technique. This technique has been described in detail before (Ranganathan, 2001; Cossaboom and Lister, 2005; Srisukvatananan, 2005).

The crystal structure and morphology of the particles were investigated using X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). The existence of strong and sharp diffraction peaks indicated the presence of the trevorite phase (isometric-hexoctahedral), NiFe_2O_4 . The lattice parameter fell between the lattice parameter of magnetite (8.339 Å) and nickel ferrite (8.396 Å). The particles were roughly spherical within the size range of 0.5–0.8 μm . Energy Dispersive X-Ray analysis (EDX) was conducted to determine the stoichiometry. Results confirmed nickel ferrite particles of composition $\text{Ni}_x\text{Fe}_{3-x}\text{O}_4$, where $0.48 \leq x \leq 0.56$. A zeta-meter indicated that the Point of Zero Charge (PZC) was at a pH of 4.7 at room temperature. Similar measurements on particles of the Alloy-800 tubing indicated a PZC of 4.4.

Fouling Loop Configuration

The loop test section is a vertical 1.5 m-long Pyrex glass column with an inner diameter of 9.93 cm with stainless steel gasketed plate closures at the top and the bottom. The top plate contains two 0.95 cm outlet ports and a 2.54 cm port for the insertion and removal of the test heater. A type-K thermocouple is mounted on the top plate for coolant temperature measurement close to the test section outlet. The test heater comprises a section of Alloy-800 steam generator tube, 50.8 cm long and 1.27 cm-diameter, containing an electrical heating cartridge. The heater can generate a maximum heat flux of 140 kW/m^2 (at the Alloy-800 surface). To ensure a smooth up-flow around the heater tube, the leading end (“nose”) is made of a plug of type 304 stainless steel machined with a parabolic profile

and brazed into position. To eliminate the entrance effect from the 1.27 cm-diameter stainless steel pipe at the bottom of the column, a 30 cm-long flow straightener consisting of a number of vertical vanes is installed.

The loop contains two 180-L tanks for coolant storage, each equipped with a 9-kW Caloritec heater (see Figure 5). Each tank is assembled with a mechanical stirrer and a nitrogen gas purging system to ensure uniform mixing and temperature distribution and an oxygen-free atmosphere. A small cooler is installed after the test section to remove heat generated by the Alloy-800 heater tube. A stainless steel centrifugal pump provides the recirculating flow.

Sample lines are located at the top and bottom of the column. These lines are used to draw samples in order to check and maintain the concentration of nickel ferrite in the loop.

Non-concomitant Removal Rate Experiments

Two preliminary runs were done to commission the loop and refine the technique of deposit measurement. After that, two runs under nominally the same operating conditions were conducted to examine the non-concomitant or direct removal (i.e., without simultaneous deposition) of deposited nickel ferrite particles into the flowing coolant. The two 180-L tanks were filled with deionized water at atmospheric pressure and 90°C and nickel ferrite was added to one to a concentration of 0.005 kg/m^3 . The pH was maintained at 9.5–10.5 by the addition of dilute potassium hydroxide solution. Measurements with the zeta meter on particles of Alloy-800 abraded off a tube sample indicated that the PZC was 4.5. The coolant pH therefore ensured that the particle transport was nominally under surface attachment control (since both the nickel ferrite and Alloy-800 would assume negative charges as their PZCs are higher than 4.5). Coolant was pumped from the first tank at a maximum rate of 12 L/min, giving an approximate Reynolds number of 10,000 in the annulus around the Alloy-800 tube. The second tank without nickel ferrite particles was by-passed for the first part of the experiment.

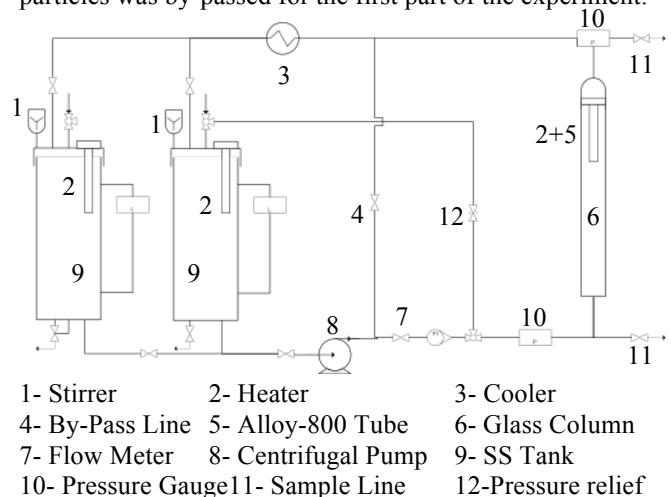


Fig. 5 Schematic diagram of the fouling loop.

Air was excluded from the system via continuous purging of the tanks with nitrogen for a minimum of 24 hours prior to the addition of nickel ferrite, and continued

throughout the experiment. To ensure consistent coolant chemistry during an experiment, a sample of coolant was taken every twelve hours via a sample line and analyzed for pH and nickel ferrite concentration. Upon stabilization of the concentration, temperature and pH, the test section was by-passed to allow for the insertion of the Alloy-800 tube. Alloy-800 bands of the same diameter as the Alloy-800 tube (1.27 cm) and 1-cm-wide were sprung onto the tube, at the bottom (near the nose), middle and top, and later removed for deposit morphology analysis (see Figure 6). The tube was introduced into the test section, switched on, and a heat flux of 112 kW/m^2 applied with a bulk coolant temperature of 90°C to induce subcooled boiling as indicated by the Jens and Lottes correlation (Jens and Lottes, 1951).

The experiment was begun for the desired exposure time. Lister and Cussac's mathematical model (2009) considered the effect of time on the deposition of magnetite in such a loop under different boiling regimes. It suggested that for 0.005 kg/m^3 bulk concentration under subcooled boiling, the deposition rate decreases for the first 145 hours and attains a steady value. In these experiments, a 170-hour exposure time was chosen to observe the release behavior after the steady-state deposition had been attained.

After 170 hours of exposure, the Alloy-800 tube was removed and the sample bands detached for examination and deposit analysis. During the next few hours, the tube was kept out of the test section, allowing deionized water to run through the loop to flush and dilute the remaining particles. Before re-installation of the tube, the coolant was sampled to determine residual nickel ferrite concentration. This was used to correct the estimate of the amount of released nickel ferrite at the end of the experiment.

During the re-exposure of the Alloy-800 tube to coolant without nickel ferrite, it was removed every two hours, for a total of 22 hours, to measure of the amount of deposit remaining.

To measure the amount of deposit, a glass rod fitted with a rubber end was used to strip the deposit off specific areas on the Alloy-800 tube adjacent to the bands. A masking tool was used to ensure that only the specific area was stripped. It consisted of a sheet of Teflon with a 1-cm by 2-cm rectangular window cut in the middle. The stripped deposit was washed off the rubber-end with HCl, and when dissolved was analysed using AA. The morphology and phase of the nickel ferrite deposited on the Alloy-800 bands after the first 170 hours were investigated using SEM and laser-Raman spectroscopy.

EXPERIMENTAL RESULTS AND DISCUSSION

Under these experimental conditions, three zones of different heat transfer characteristics were observed. As the fluid flows upwards along the vertical tube, the surface and fluid temperatures at the interface vary. The locations of the Alloy-800 bands are seen in Figure 6.

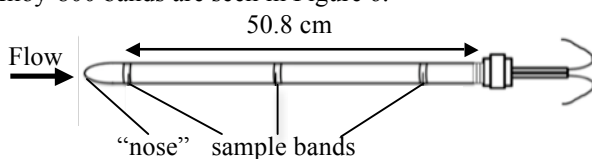


Fig. 6 Schematic diagram of the Alloy-800 heater tube.

Since, a steam gap was occasionally observed between the heater and the sample bands, the deposit on the bands may not have been a consistently accurate reflection of the morphology on the heater. However, Figure 7 shows the evolution of nickel ferrite deposition along the tube and there are similarities between the direct deposition on the tube and the deposition on the sample bands. Presumably, a steam gap contributed to non-uniform deposition on the sample bands.

Near the 'nose' of the heater tube, the fluid temperature at the metal surface is close to the inlet temperature. As the fluid flows along the tube, its temperature at the surface increases and the thermal boundary layer thickens, thus enhancing bubble nucleation. Relatively stagnant bubbles with large diameters and roughly spherical in shape were formed on the sample band near the nose. From the middle to the top section of the tube, bubbles were smaller in diameter. They nucleated very rapidly and appeared to move along the tube before collapsing. The deposit characteristics were also different along the tube length.

As illustrated in Figure 8, at the start of the boiling zone close to the nose of the tube the deposition had a pattern of small rings of diameter about 0.4 mm. Further along the tube, where the surface temperature was higher, small clumps of particles were observed (Figure 9).

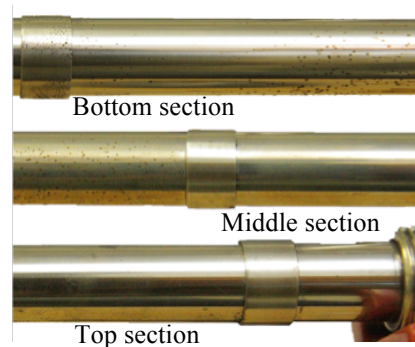


Fig.7 Nickel ferrite deposition under three different heat transfer characteristics on Alloy-800 tube and sample bands.

Observations of the tube surface during subcooled boiling revealed a strong influence of bubble behavior on the deposition process. The rings or spots of deposit were associated with bubble nucleation sites, as observed by Basset et al. (2000) in the deposition of magnetite on Alloy-800 heat exchange surfaces under subcooled boiling. Particle agglomeration was observed in the deposit rings as shown in Figure 10. This observation suggested that particles are trapped from the bulk liquid and swept to the surface by the disturbed vapour-liquid interface of growing bubbles.

It is postulated that some of the already-formed deposit is removed by the nucleating and growing bubbles by trapping at the vapour-liquid interface by stable bubbles and bubbles collapsed on or close to the surface. A less well-defined distribution of bubbles tended to form near the downstream end of the test section, where bubble nucleation was more frequent and bubble diameters were smaller.

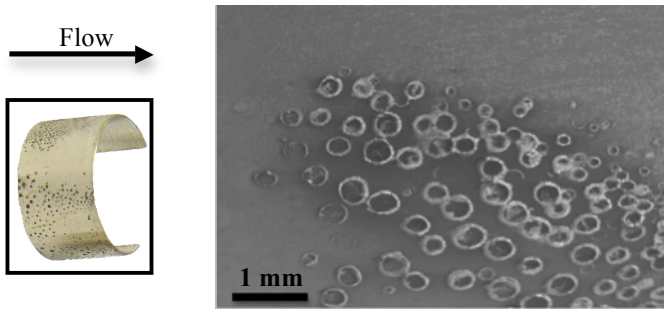


Fig. 8 SEM picture of nickel ferrite deposition at a high degree of subcooling close to the heater nose.

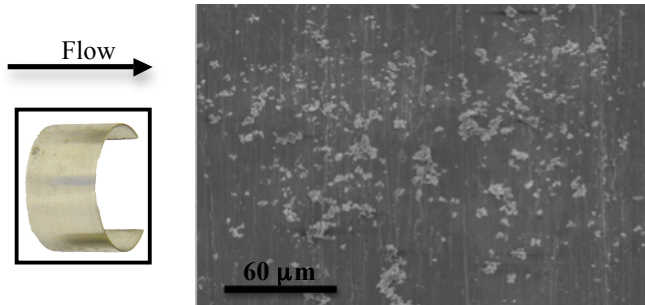


Fig. 9 SEM picture of nickel ferrite deposition at a low degree of subcooling close to the top of the heater.

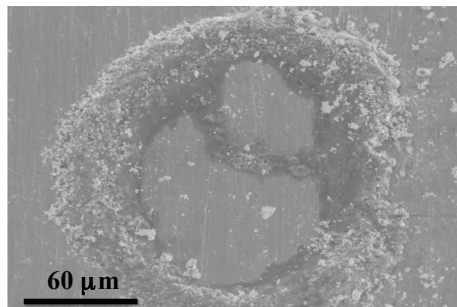


Fig. 10 Nickel ferrite particles agglomerated on a bubble nucleation site.

In each run, samples from three sections along the heater length were taken for analysis after the 170 h of exposure. The measurements of nickel ferrite concentration from both runs showed that the heaviest deposit was in the region at the heater nose (see Figures 11 and 12). The release curves in the figures are assumed exponentials, each with its removal constant.

Make-up nickel ferrite was added to maintain C_b at the desired value. However, variations occurred and average C_b was different in run 1 and run 2. The higher average C_b in run 1 gave a higher deposit mass at the end of the deposition period (170 h). The calculation of deposition velocity normalises for concentration by dividing it into the deposition at 170 h. The values of K_d for the two runs are comparable, as shown by the variance in Table 2.

Experimental results are compared to the particulate fouling model in prediction Table 2. The theoretical value is derived from the predicted amount of nickel ferrite deposition on the middle part of the heater. The amount of

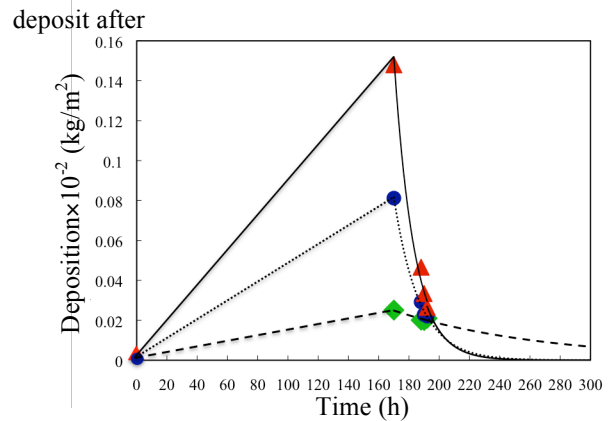


Fig. 11 Nickel ferrite deposition on Alloy-800 tube for the first 170 hours at average $C_b = 7.5\text{ppm}$ followed by exposure to particle-free coolant from run 1 (tube bottom (nose) = $\blacktriangle$, middle = $\bullet$, top = $\blacklozenge$).

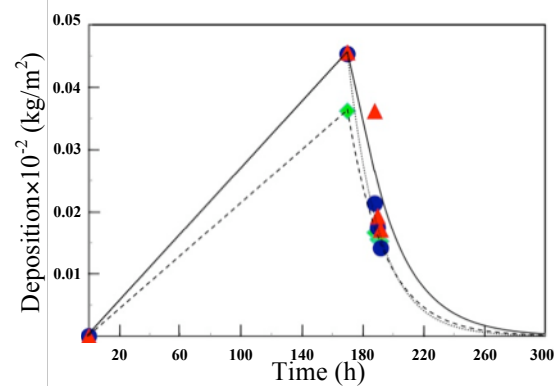


Fig. 12 Nickel ferrite deposition on Alloy-800 tube for the first 170 hours at average $C_b = 5.9\text{ppm}$ followed by exposure to particle-free coolant from run 2 (tube bottom (nose) = $\blacktriangle$, middle = $\bullet$, top = $\blacklozenge$).

170 hours averaged for the two runs is $6.33 \times 10^{-4} \text{ kg/m}^2$, about 97% of the predicted value ($6.53 \times 10^{-4} \text{ kg/m}^2$). The prediction is in good agreement with the results.

Table 2. Average deposition velocity (K_d) at each location on the heater tube compared to the theoretical value from the particulate fouling model (Lister and Cussac, 2009).

	Section on tube		
	Top	Middle	Bottom
average K_d ($\mu\text{m/s}$)	0.100 ± 0.018	0.207 ± 0.059	0.316 ± 0.169
theoretical K_d ($\mu\text{m/s}$)	-	0.213	-

The removal constants (k_{r0}) were obtained from the slopes of the release curves at 170 h. The values are presented in Table 3. From the model, k_r ($2.31 \times 10^{-6} \text{ s}^{-1}$) is the removal constant when the deposition and removal processes take place simultaneously. The measured k_{r0} ($9.27 \times 10^{-6} \pm 1.09 \times 10^{-6} \text{ s}^{-1}$), however, is when the source term is removed from the coolant and there is no deposition. The difference may well reflect the competing mechanisms of deposition and removal accounted for in the model.

The nickel ferrite deposition decreases along the length of the heater. Two factors may account for this variation. They are the depletion of source term and the boiling intensity along the heater length. When the deposition takes place continuously, the nickel ferrite concentration in the coolant is depleted downstream from the inlet condition (nominal 5ppm). The boiling parameter is computed from Equation (6). Under the same heat flux, the boiling parameter increases with T_{film} , which is expected to increase from the heater nose to the top of the column. The boiling parameter has an effect on the amount of deposition.

Table 3. Removal constant (k_{r0}) at each location on the heater tube compared to the theoretical value for release with concomitant deposition (Lister and Cussac, 2009).

	Section on tube		
	Top	Middle	Bottom
average k_{r0} (s^{-1})	6.19×10^{-6} $\pm 3.12 \times 10^{-6}$	9.27×10^{-6} $\pm 1.09 \times 10^{-6}$	8.83×10^{-6} $\pm 5.65 \times 10^{-6}$
theoretical k_r (s^{-1})	-	2.31×10^{-6}	-

The present experiments led to plots showing release without concomitant deposition. Previous experiments had exhibited consolidation, where the unconsolidated portion of the deposit was approximately 80%. All of the deposits in Figures 11 and 12 released material and tended towards zero, however.

Coolant pH modifies the characteristics of the surface charges, the electrical double layer and the London-Vander-Waals interactions, and therefore affects deposition. The present experiments were done with a measured coolant pH of 9.5-10.5, which is greater than the PZCs of the nickel ferrite particles and the Alloy-800. Both surfaces should therefore have negative charges, making the deposition attachment-controlled. It is remarkable, then, that quantitatively the agreement with the predictions is good, since the model was derived for transport control. One difference arising from the control regime may be the fact that the deposits in the experiments reported here exhibited no consolidation.

CLOSING REMARKS

The particulate fouling mathematical model as proposed by Lister and Cussac (2009) is in reasonable agreement with previous experimental work. The model predicts quite well experimental results under isothermal conditions, non-boiling conditions and subcooled boiling conditions. Under bulk boiling conditions, the model over-predicts the experimental data probably because of underestimating fluid disturbances caused by bubble formation and detachment.

A set of experiments was done to extend the understanding of removal and consolidation rates of nickel ferrite under subcooled boiling conditions. It was found that, under nominal attachment control, deposition rates were similar to those, found previously under transport control. However, the deposits appeared to decline to zero after a certain period of time, in contrast to those under

transport control. Further studies to validate these results and to obtain similar data under a range of conditions would be valuable.

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NOMENCLATURE

BR	boiling enhancement parameter, dimensionless
C_b	concentration in the bulk, kg/m^3
d_p	particle diameter, m
d_p^+	dimensionless particle diameter ($=d_p u^*/\nu$)
f	friction factor ($=0.791/Re^{0.25}$)
k_c	consolidation constant, s^{-1}
k_r	removal constant, s^{-1}
k_{r0}	non-concomitant removal constant, s^{-1}
K_d	deposition coefficient in single phase, m/s
K_{diff}	diffusion coefficient, m/s
K_{th}	thermophoretic velocity, m/s
L	latent heat of vaporisation, J/kg
m_f	mass of fouling, kg
m_{labile}	labile parameter, dimensionless
q	heat flux, kW/m^2
R_b	bubble radius, m
Re	Reynolds number, dimensionless
Sc	Schmidt number, dimensionless
t	time, s
t_c	critical time to start the consolidation, s
T_{film}	temperature of film, K
T_{sat}	saturation temperature, K
u	flow velocity in the bulk, m/s
u^*	friction velocity, m/s
λ	thermal conductivity, W/mK
ν	kinematic viscosity, m^2/s
Φ_d	particle deposition flux, $kg/m^2.s$
Φ_r	particle removal flux, $kg/m^2.s$

Subscript

p	particle
w	water

REFERENCES

- Asakura, Y., et al., 1978, Deposition of iron oxide on heated surfaces in boiling water, *Nuclear Science and Engineering*, V.67, N.1, pp.1-7.
- Basset, M., McInerney, J., Arbeau, N., and Lister, D. H., 2000, The fouling of Alloy-800 heat exchange surfaces by magnetite particles, *The Canadian Journal of Chemical Engineering*, V. 78, pp. 40-52.
- Callamand, S. P., 1999, Numerical simulation of corrosion product deposition on heat exchanger surfaces, *MSc. Thesis*, Department of Chemical Engineering, University of New Brunswick, Canada.
- Capentier, H., 2002, Numerical simulation of corrosion product deposition onto heat exchange surfaces in sub-cooled boiling conditions, *MSc. Thesis*, Department of

Chemical Engineering, University of New Brunswick, Canada.

Cleaver, J. W. and Yates, B., 1975, A sublayer model for the deposition of particles from a turbulent flow, *Chem. Eng. Sci.*, Vol. 30, pp. 983-992.

Cossaboom, J. L. and Lister, D. H., 2005, The fouling of Alloy 800 heat exchanger tubes by nickel ferrite under bulk boiling conditions, ECI Symposium Series, *Proc. 6th Int. Conf. Heat Exchanger Fouling and Cleaning*, Kloster Irsee, Germany, Vol. RP2, pp. 109-118.

Epstein, N., 1988, Particulate fouling of heat transfer surfaces: mechanisms and models, fouling science and technology, L.F. Melo et al. ed., *Kluwer Academic Publishers*, pp. 143-164.

Epstein, N., 1997, Elements of particle deposition onto nonporous solid surfaces parallel to suspension flows, *Experimental Thermal and Fluid Science, Elsevier Science Inc.*, Vol. 14, pp. 323-334.

Gudmundsson, J. S., 1981, Particulate fouling, *Proc. Int. Conf. Fouling of Heat Transfer Equipment (Rensselaer Polytechnic Institute-August 1979)*, Heat Transfer Equipment, E. F. C. Somerscales, J. G. Knudsen, Hemisphere Publishing Corporation, pp. 357-387.

Jens W. H. and Lottes P. A., 1951, Analysis of heat transfer, burnout, pressure drop and density data for high-pressure water, ANL-4627.

Kern, D. Q. and Seaton, R. E., 1959, A theoretical analysis of thermal surface fouling, *British Chemical Engineering*, Vol. 4, pp. 258-262.

Lahey, R. T., 1992, Boiling heat transfer: modern developments and advances, *Elsevier Science Publishers B.V.*

Lister, D. H. and Cussac, F., 2009, Modelling of particulate fouling on heat exchanger surfaces: influence of bubbles on iron oxide deposition, *Heat Transfer Engineering*, Vol. 30, pp. 840-850.

Lister, D. H. and Venkateswaran, G., 1995, Corrosion products in nuclear reactor coolants. *Proc. First National Conference on Corrosion and its Control*, Indian institute of technology, Bombay, India.

Margotin, J. P., 1994, Simulation of corrosion product transport in flowing water, *MSc. Thesis*, Department of Chemical Engineering, University of New Brunswick, Canada.

McCrea, L., 2001, Deposition of corrosion product particles onto heat exchange surfaces, *MSc. Thesis*, Department of Chemical Engineering, University of New Brunswick, Canada.

McNab G. S. and Meisen A., 1973, Thermophoresis in liquids, *Journal of Colloidal and Interface Science*, Vol. 44, pp. 339-346.

Turner, C. W. and Klimas, S. J., 2001, The effect of surface chemistry on particulate fouling under flow-boiling conditions, *Proc. Eng. Foundation Conf. Heat Exchanger Fouling*, Davos, Switzerland, pp. 19-26.

Ranganathan, S., 2001, Model corrosion products of relevance to water-cooled nuclear reactors: characterisation and solubilisation studies, *Ph.D. Thesis*, Department of Chemistry, University of Madras.

Srisukvatananan, P., 2005, The deposition of nickel ferrite onto heated Zircaloy-4 surfaces in high-temperature boiling water: interactions with dissolved zinc and boron, *MSc. Thesis*, Petroleum and Petrochemical College, Chulalongkorn University, Thailand.

Yung, B. P. K., et al., 1989, The role of turbulent bursts in particle re-entrainment in aqueous systems, *Chemical Engineering Science*, Vol. 44, pp. 873-882.