## THE EFFECT OF THE REMOVAL OF STEAM GENERATOR TUBE ID DEPOSITS ON HEAT TRANSFER

S. J. Klimas, D. G. Miller, J. Semmler, and C.W. Turner

### ABSTRACT

The thermal resistance of boiler primary-side tube deposits from the Gentilly-2 NGS (Hydro-Québec) was evaluated by an experimental comparison of the heat transfer rates between fouled samples and identical, factory-new, "clean" tubing. The deposits were subsequently removed using either a chemical decontamination process (CAN-DEREM<sup>TM</sup> Plus) or a mechanical cleaning process (Siemens SIVABLAST<sup>TM</sup>) in two stages. After each removal, the thermal resistance of the remaining deposit was re-measured.

The 90- to 150- $\mu$ m-thick deposits on the inside diameter of steam generator cold-leg tubes were found to pose significant resistance to heat transfer (0.05 to 0.06 m<sup>2</sup>·K/kW at 210°C). However, the 10- to 30- $\mu$ m-thick dense layers remaining on the tubes after the decontamination were found to have no measurable effect on the heat transfer. The thin, 2- $\mu$ m, tube deposit on the steam generator hot leg slightly enhanced heat transfer.

The measured thermal resistance results in a calculated thermal conductivity of 1.5 W/m·K for the 90-µm-thick deposit. The 150-µm-thick deposits were found to consist of two layers: an outer surface layer having an average porosity of 50% and a conductivity of 2.3 W/m·K, and an inner layer with an average porosity of 5% and a conductivity of more than 3.0 W/m·K.

The previous best estimate of the thermal conductivity was 1.4 W/m·K for the porous magnetite deposits that had formed on the primary side of nuclear steam generators with thickness <90  $\mu$ m. This work confirms this number but also demonstrates that it is applicable only for porous, unconsolidated deposits. The conductivity increases for thicker deposits because of increasing deposit consolidation, particularly at the most inner layer adjacent to the tube metal.

Atomic Energy of Canada Ltd. Chalk River Laboratories Chalk River, Ontario KOJ 1J0 Canada

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# INTRODUCTION AND OBJECTIVE

This work was initiated for purely practical reasons—the utility (Hydro-Québec) wished to predict the degree of recovery of the steam generator thermal performance if a cleaning of the primary heat transport side of their boilers was undertaken. The cleaning of the boiler primary side is conducted mostly to reduce the radiation fields, but the recovery of thermal performance is also a major consideration.

The degradation of thermal performance is a sum of several components, major of which are the primary-side fouling, the secondary-side fouling, and the divider plate leakage. These measurements isolate the effect of the primary-side fouling.

# DESCRIPTION OF THE EXPERIMENTAL METHOD

## Workscope

The workscope consisted of several steps to characterize the tube deposit and to measure its thermal resistance. Details are presented in Table 1.

Step	Samples 1 and 2	Samples 3 and 4
1	Measure the Deposit Thermal Resistance	Measure the Deposit Thermal Resistance
2	Determine the Surface Roughness and the	Determine the Surface Roughness and the Deposit Thickness
	Deposit Thickness	and Loading
3	Apply the Chemical Cleaning Process	Apply the Mechanical Cleaning Process for a Very Short
		Time
4	Determine the Amount of the Deposit	Measure the Thermal Resistance of the Remaining Deposit
	Removed in Step 3	
5	Measure the Thermal Resistance of the	Determine the Surface Roughness and the Thickness and
	Remaining Deposit	Loading of the Remaining Deposit
6		Apply the Complete Mechanical Cleaning Process
		Measure the Thermal Resistance of the Remaining Deposit
7		Determine the Surface Roughness and the Thickness and
		Loading of the Remaining Deposit.

## Table 1: The outline of the Experimental Workscope

Four samples were submitted for the investigation. One sample (Sample 1) was obtained from the hot leg, three other samples (Samples 2, 3 and 4) originated at the cold leg. Details are given in Table 2. Samples 1 and 2 were processed together. Their thermal resistance was measured twice; before and after the chemical decontamination. In a separate program, Samples 3 and 4 were processed (also simultaneously). This time, the thermal resistance was measured three times, the samples being cleaned in between measurements using a mechanical process.

Sample Name	Sample Description
Sample 1	Deposits on the Tube Internal Diameter, Sample from Gentilly-2 NGS, Steam Generator 2,
	Hot Leg Below the Second Support Plate
Sample 2	Deposits on the Tube Internal Diameter, Sample from Gentilly-2 NGS, Steam Generator 2,
_	Cold Leg, Below the Third Baffle Plate in the Preheater
Sample 3	Deposits on the Tube Internal Diameter, Sample from Gentilly-2 NGS, Steam Generator 2,
_	Cold Leg, Below the Third Baffle Plate in the Preheater, Immediately Above Sample 2
Sample 4	Deposits on the Tube Internal Diameter, Sample from Gentilly-2 NGS, Steam Generator 2,
	Cold Leg, Below the Third Baffle Plate in the Preheater, Immediately Above Sample 3
Reference Tubing	Factory-New 5/8" (15.875 mm) OD x 0.0445" (1.13 mm) Wall Incoloy Alloy 800 Tubing

Table 2: List of the Samples

### **Determination of the Thermal Resistance of Deposits**

The deposit was located on the internal surface of a 5/8" OD tubing. Therefore, the tubing sample was mounted in an experimental loop so that high-temperature high-pressure water could be recirculated inside the tube. The tube was directly electrically heated using a low-voltage, high-current power supply. The schematic of the test section is presented in Figure 1.



Figure 1: The experimental test section.

The water flow rate and the tube heating rate were matched so that single-phase forcedconvective heat transfer conditions were maintained, i.e., no boiling was occurring. A detailed description of the experimental method is provided by Turner et al. (1998). The relevant experimental conditions are listed in Table 3, and the stability of the experimental conditions is illustrated in Figure 2.

Flow Conditions	Vertical Up-flow of Water Inside a Tube	
Heat Transfer Mode	Forced-Convection to Single-Phase Water	
Method of Heating	60 Hz Low-Voltage Direct Electrical Heating	
Pressure	6.1 MPa	
Film Temperature Range	130 to 210°C	
Heat Flux	70 to 170 kW/m <sup>2</sup>	
Mass Flux	400 kg/m <sup>2</sup> ·s	
Reynolds Number	26 000 42 000	

#### Table 3: The Experimental Conditions



Figure 2: Stability of the experimental conditions during the tests.

The measured parameters are the heat input (electrical), the water flow rate, the bulk temperature at the inlet to the test section, the temperature on the outlet from the test section, and several temperatures of the tube wall.

The temperature difference that drives the flow of heat from the tube wall into the flowing water is evaluated from these experimental data. The measurements were performed under similar conditions for the fouled sample and for a length of identical "clean" reference tubing.

The thermal resistance of deposits is evaluated from

$$R_{d} = \left[\frac{T_{\text{wall}} - T_{b}}{q^{''}}\right]_{\text{fould}} - \left[\frac{T_{\text{wall}} - T_{b}}{q^{''}}\right]_{\text{clean}}.$$

The deposit thermal conductivity is calculated from

$$\kappa_{d} = \frac{\delta}{R_{d}}.$$

#### **Oxide Removal and Characterization**

The deposit is almost pure magnetite. It precipitates from solution as it traverses the steam generator as a consequence of the normal solubility curve of magnetite. The contribution of particulate fouling is thought to be significant in some instances.

Two proprietary methods for cleaning of nuclear steam generators were used for this laboratoryscale trial. CAN-DEREM<sup>TM</sup> Plus process (Miller et al., 1997) was used for decontamination of Samples 1 and 2. CAN-DEREM<sup>TM</sup> Plus is a chemical decontamination process employing EDTA/citric acid under reducing conditions. Siemens SIVABLAST<sup>TM</sup> was used for mechanical cleaning of Samples 3 and 4 in two stages to differentiate between the outer and inner layer of the deposit. SIVABLAST<sup>TM</sup> is a mechanical process that involves blasting the tube wall with stainless steel shot. The description of these methods is outside of the scope of this paper.

The deposit loading [kg of Fe<sub>3</sub>O<sub>4</sub> per m<sup>2</sup> of tube surface] was determined by descaling of the surface with alkaline permanganate solution (3% KMnO<sub>4</sub> and 7% KOH) and an analysis of the solution for iron. The deposit thickness was determined from scanning electron microscopy (SEM) micrographs of metallographic cross sections. SEM was also used to determine the morphology of the deposits to help distinguish between the possible fouling mechanisms that created the deposits. The deposit porosity was calculated from deposit loading, its thickness, and the known literature density of magnetite.

### RESULTS

Figures 3 (a) and (b) show the changes in film resistance with film temperature for fouled and clean tubes before and after chemical cleaning. The distance between the lines obtained for "clean" and "fouled" tubes is interpreted as the thermal resistance of the deposit. Figure 3 (a) shows that the film resistance of the cold leg sample is higher than the film resistance for the reference clean tube. On the other hand, the hot leg sample has a slightly lower film resistance than the clean tube does. After the chemical cleaning, Figure 3 (b), all the lines overlap showing that the film resistance is virtually identical.



Figure 3: Examples of experimental curves: (a) before chemical decontamination and (b) after the chemical decontamination.

The summary of the results is presented in Table 4.

	Sample	Deposit	Deposit	Surface	Deposit thermal
		Thickness,	Porosity,	Roughness	Resistance Measured
		μm	%	$, R_a, \mu m$	at 210°C, m <sup>2</sup> ·K/kW
Sample 1	"As-Received"	1.8	-	1.0	-0.012
	After Chemical Cleaning	0.7	-	0.7	0.005
Sample 2	"As-Received"	90	53	4.6	0.059
	After Chemical Cleaning	30	-	2.5	0.011
		2.0		0.5	
Sample 3	"As-Received"	155	38	3.3	0.05
	After the First SIVABLAST <sup>TM</sup>	24	4	1.8	-0.01
1	Cleaning Step				
	After the Second SIVABLAST <sup>TM</sup>	8.0	6	1.2	0.00
	Cleaning Step				
Sample 4	"As-Received"	150	43	3.3	0.06
	After the First SIVABLAST <sup>TM</sup>	33	8	2.0	0.00
	Cleaning Step				
	After the Second SIVABLAST <sup>TM</sup>	9.7	3	1.3	-0.01
	Cleaning Step	4			
Reference	Tubing	0.0	-	0.9	0.00

## Table 4: Summary of the Experimental Results

The thin deposits on hot leg, Sample 1, slightly enhanced the heat transfer ( $R_d = -0.01 \text{ m}^2 \cdot \text{K/kW}$ ). After chemical cleaning, the measured resistance was zero within the experimental uncertainty.

The 90- to 150- $\mu$ m-thick tube deposits on all cold leg samples (Samples 2, 3 and 4) posed significant resistance to heat transfer (0.05 to 0.06 m<sup>2</sup>·K/kW at 210°C). At the same time, the 10- to 30- $\mu$ m-thick dense layers remaining on the tubes after the cleaning were found to have no measurable effect on the heat transfer.

The experimental uncertainty is estimated to be 0.01 m<sup>2</sup>·K/kW for Samples 1 and 2, and 0.015 m<sup>2</sup>·K/kW for Samples 3 and 4.

# DISCUSSION

Comparison of the deposit porosity data in Table 3 leads to the conclusion that the deposits on Samples 3 and 4 have a two-layer structure: a dense inner layer and a more porous outer layer. (This is analogous to the secondary-side deposits, which are also found to have a layered structure [Turner et al., 1998].) For these samples, using the data from Table 4 and assuming the two-layer model, the calculated porosity of the deposit outer layer is 46 and 53% respectively (average 50%), whereas the average porosity of the inner layer was 5%. The two layers have distinctively different mechanical and thermal properties: the outer layer is much softer and relatively non-conductive, whereas the inner layer is hard (more difficult to remove) and more thermally conductive.

Sample 2 appears to have consisted largely of a single layer of deposit material with a low thermal conductivity, although there is evidence of "islands" of hard-to-dissolve deposit.

Table 5 presents thermal conductivity of the deposits calculated on the basis of the measured deposit thermal resistance, thickness, and the interpretation of the data.

Table 5: Thermal Conductivity of Tube Deposits,  $\kappa_d$ , Calculated on the Basis of the Experimental Results and their Interpretation

D II			
Deposit	δ, μm	Interpretation	$\kappa_d$ at 210°C, W/m·K
Deposit on Sample 1	2	Increased Surface Roughness	Equivalent Thermal
		, i i i i i i i i i i i i i i i i i i i	Resistance of -0.01
			m².K/kW
Entire Deposit on Sample 2	90	Porous Deposit, Porosity of 53%	1.5
Deposit on Samples 3 and 4 which was	120	Porous Outer Layer of the	2.3
Removed in the First Cleaning Step		Deposit, Average Porosity of	
		50%	
Deposit on Samples 3 and 4 Remaining	30	Deposit Inner Dense Layer,	>3
after the First Cleaning Step		Porosity of 5%	

<sup>\*</sup> The thermal conductivity of pure substances at 210°C is 0.67 W/m·K for water (ASME Steam Tables) and 3.6 W/m·K for magnetite (Mølgard and Smeltzer, 1971).

The experimental results are reported at a film temperature of 210°C. Extrapolation to 270°C, a temperature more relevant to the steam generator conditions, was also conducted and the thermal conductivity of the deposits at 270°C was predicted to be lower by approximately 15%. (This decrease exceeds the drop in the thermal conductivity of pure magnetite and water in this temperature range.)

Deposits increase the surface roughness, which may result in an equivalent thermal resistance of up to  $-0.01 \text{ m}^2 \text{ K/kW}$  (the negative sign indicates that the heat transfer is enhanced). A relationship that is often observed is that the thicker the deposit, the higher the surface roughness. This proved to hold in this investigation.

Figures 4 (a), (b) and (c) show typical SEM micrographs of the metallographic cross sections. They are consistent with the presented interpretations. Figure 4 (a) shows a thick deposit before mechanical removal (typical view of Samples 3 and 4). The deposit thickness is relatively uniform and visibly porous.

Our experience indicates that deposits of irregular thickness tend to be produced when the growth rate is high, or when existing deposits are subjected to aggressive dissolution. Figure 4 (b) demonstrates that this was the case for chemically cleaned sample (Sample 2). The irregularity of the deposit thickness reduces the "effective thickness" of the deposit, and is therefore beneficial from the point of view of thermal performance. (The "effective thickness", for heat transfer purposes, is the harmonic mean thickness of the deposit, which is always smaller than the arithmetic mean.) This effect might be at least partly responsible for the low resistance of the deposit remaining after the chemical cleaning. The deposit has the appearance of being

even more porous than the deposit on the samples that were subject to mechanical cleaning (Samples 3 and 4). This is consistent with its lower conductivity.

Figures 4 (c) and (d) show typical views of the deposit after the first and second mechanical cleaning steps (Sample 3 and 4). The deposit thickness is uniform, and it appears to be well consolidated. The darker thin layer adjacent to the metal surface can be interpreted as the intrinsic corrosion layer on the metal. The balance of the deposit can be interpreted as the layer produced during the hot conditioning performed during the station commissioning. There are also fouling mechanisms that predict increased deposit consolidated near the wall before the mechanical removal was applied. However, the possibility that the mechanical cleaning process itself compacted the deposit at the wall cannot be totally excluded.



Figure 4: Examples of SEM micrograph of metallographic cross sections showing the thickness profile of the tube deposits (a) before removal, (b) after chemical removal, (c) after mechanical removal (first step), and (d) after the mechanical removal (second step).

The thermal conductivity of primary-side tube deposits from CANDU<sup>®</sup> stations were measured previously using a similar experimental technique (Turner and Klimas, 1994; Turner et al.,

1998). The best estimate to date of the thermal conductivity of CANDU deposits is 1.4 W/m·K and the current investigation confirms this number but it also indicates that it is only applicable for porous, unconsolidated deposits. The thermal conductivity of thicker (over 100  $\mu$ m) and less porous deposits can be much higher. A short interpretation of this difference is given below, and a wider review is presented in Turner et al., 1998.

Euler (1957), identifies a class of the physical properties of composite materials whose value depends on the arrangement of the constituting phases in the sense defined by the Maxwell equation for the over-all conductivity, i.e., which depend on flux or force transmission through the phases. Thermal conductivity belongs to this class. There are two limiting cases: isolated pores in a continuous solid matrix, and solid grains mixed with continuous voids. (For steam generator primary-side deposits, the solid is magnetite, the void is water.) The Maxwell equation (or any other of the numerous existing models) can be used for either limiting case. However, these two cases do not converge for intermediate porosity.

According to Rice (1995), there is a discontinuity in the properties of ceramic materials at the "percolation limit". (The percolation limit is a critical porosity at which the structure of the solid becomes fragmented.) This discontinuity occurs because the effective thermal conductivity is determined by the minimum solid area (MSA), and not directly by the porosity.

Consequently, the thermal conductivity of magnetite-water composite has to be described by at least two different formulas: one for "unconsolidated" and the other for "consolidated" deposit. A schematic of such a model is presented in Figure 5. Assuming a deposit of 50% porosity, the thermal conductivity of an unconsolidated deposit is expected to be approximately 1.5 W/m·K at 210°C, whereas the conductivity of consolidated deposits is expected to be between 2.0 to 3.6 W/m·K, depending on their porosity. At 270°C, the model predicts the conductivity of unconsolidated deposit to be 1.4 W/m·K.



Figure 5: Schematic representation of the dependence of the thermal conductivity of a magnetite-water composite on porosity (volume fraction of water).  $\Phi_c$  is the percolation porosity of the structure at which the particles of magnetite become effectively separated from each other.

# IMPLICATIONS AND CONCLUSIONS

The deterioration of the thermal performance in CANDU (CANada Deuterium Uranium) stations is manifested by an increase in the reactor inlet header temperature (RIHT). The THIRST thermal-hydraulic code predicts that the measured primary-side deposit thermal resistance would increase the reactor inlet header temperature by up to 3.8°C. The removal of 70% of this deposit would recover up to 2.5°C of the RIHT.

In conclusions:

- Primary-side fouling may constitute a significant fraction of the thermal performance loss observed in CANDU<sup>®</sup> stations.
- The thermal conductivity of CANDU steam generator primary-side deposits is 1.4 W/m·K for unconsolidated deposits, and more than 3.0 W/m·K for consolidated deposits.
- Chemical or mechanical cleaning is effective in restoring the thermal performance lost due to fouling of the primary side of the steam generator.

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## NOMENCLATURE

	Symbol	Definition	Unit
	R <sub>d</sub>	deposit thermal resistance	m².K/W
	T	temperature	K
	q"	heat flux	W/m²
Greek:	δ	deposit thickness	m
	Φ <sub>c</sub>	critical porosity	-
	κ	thermal conductivity	W/m∙K
Subscripts:	b clean d fouled wall	of the bulk water without the deposit of the deposit with the deposit of the tube inner wall	

### ACKNOWLEDGMENTS

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We would like to thank those participants of our team without whom the program could not have been successfully completed.

At Atomic Energy of Canada Ltd. (AECL), Mike Brideau ran the experimental high-pressure recirculating loop, Brian Barry provided chemical cleaning of the tubes and determination of the deposit loading, Frank Szostak performed scanning electron microscopy (SEM), Mark Seguin performed laser profilometry, and Roger Klatt assisted in the machine shop.

Klaus Streit of the Siemens Power Company provided laboratory-scale decontamination using the SIVABLAST<sup>™</sup> method.

Steve Plante and Claude Drouin of Hydro-Québec provided excellent guidance throughout the program.



### DISCUSSION

Authors: S.J. Klimas, D. Miller, J. Semmler, C.W. Turner, AECL

Paper: The Effect of the Removal of Steam Generator Tube Deposits on Heat Transfer

Questioner: D.H. Lister, University of New Brunswick

#### **Question/Comment:**

How was the porosity of the deposits determined? And did you consider characterizing the pore geometry? The latter point has an obvious application to thermal resistance, as we would expect from the two-layer deposit, for example, that you mention.

### **Response:**

The porosity was determined from the deposit loading and its thickness. The pore geometry was not characterized in any way other than inspection of the SEM micrographs.

## Questioner: C. Taylor, AECL

### **Question/Comment:**

You stated the benefit of removing 70% of the primary-side fouling from Gentilly2. What would happen if you removed 100%. Is it always better to remove 100% of the primary-side fouling?

#### **Response:**

Our investigation showed it is restored almost fully by removing the porous outer layer of the deposit. The dense inner layer does not have much effect on thermal resistance even if it has a thickness of 30  $\mu$ m. As a matter of fact, removal of the final few  $\mu$ m of the deposit could slightly hinder the heat transfer if it reduces the surface roughness.

However, the thermal performance is only one of many issues that drive the cleaning of the steam generator primary side. Other important issues are the radiation fields, pressure drop, and the prospect of recontamination/re-deterioration of thermal performance after the cleaning. The utility must consider them all in their analysis before the cleaning is undertaken.

### Questioner: G. White, Dominion Engineering

## **Question/Comment:**

Did you attempt to duplicate the thermal conductivity results analytically using a model of conduction through a matrix of magnetite and liquid water?

### **Response:**

There is an abundance of models published in the literature to predict the thermal conductivity of porous matrices. All of them have the form:

$$k_{\rm ee} = f(k_{\rm A}, k_{\rm B}, \varepsilon_{\rm A}, \varepsilon_{\rm B}),$$

i.e., the effective thermal conductivity of the matrix is a function of the thermal conductivity of pure components A and B, and their volumetric fractions in the matrix. In our opinion, models in this form are not generally applicable because they do not take into account the important structural effects.

We find that there is a considerable scatter of the conductivity predicted by the various models. The model that comes closest to predicting our experimental results for unconsolidated porous deposit is a simple in-series arrangement of magnetite and water. The difficulty at this point is to find a justification for using a particular model for a given station deposit.

Questioner: R.F. Voelker, Lockheed-Martin

#### **Question/Comment:**

How did you determine the fouling factor from your test measurements?

#### **Response:**

The heat flux was determined from the change in water enthalpy across the test section and the flow rate. The local temperature of the wall was determined by a direct measurement. The temperature of the bulk water was calculated from the inlet temperature and the rate of heat addition to the water. Finally the deposit thermal resistance was calculated from the equation:

$$R_{d} = \left[\frac{T_{wall} - T_{f}}{q}\right]_{fould} - \left[\frac{T_{wall} T_{f}}{q}\right]_{clean}$$