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THE EFFECT OF OXIDE FILMS ON THE THERMAL RESISTANCE BETWEEN CONTACTING ZIRCONIUM ALLOYS

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ABSTRACT

The resistance to heat flow at interfaces formed between contacting solids plays a key role in nuclear reactor systems. In many instances, these contacts occur at elevated temperatures and in oxidizing or corrosive environments. These environments normally lead to increased thermal resistance (decreased conductance), which recent theoretical predictions have related to (i) thermal conductivity of the oxide layer, (ii) layer thickness, (iii) effective hardness of the surface and (iv) surface microtopography.

Experiments with oxidized zirconium specimens have confirmed these predictions and revealed that the contact conductance in vacuum increases as the ratio of layer thickness to contact-spot radius increases.

This study is of particular importance in our efforts to predict the rate of heat flow from an overheated CANDU pressure tube when it comes into contact with the surrounding calandria tube during a postulated loss-of-coolant accident.

NOMENCLATURE

A_a, A_r	apparent and real areas of contact, m^2
a	contact-spot radius, μm
b	flux tube radius, μm
H, H_e	hardness and effective hardness, MPa
h_δ, h	thermal contact conductance, with and without layer, $W/(m^2.°C)$
k	thermal conductivity, $W/(m.°C)$
m	mean asperity slope
N	number of microcontacts
n	contact-spot density
P_a	contact pressure, MPa
Q_s	steady-state heat flow, W
R_δ, R	thermal constriction resistance, with and without layer, °C/W
ΔT	interface temperature drop, °C
t	oxide layer thickness, μm
x	$Y/\sqrt{2}\sigma$
Y	separation between mean surface planes, μm

Greek Symbols

β	conductivity parameter, $(\kappa-1)/(\kappa+1)$
γ	geometric parameter, t/b
δ	relative layer thickness, t/a
ϵ	geometric parameter, a/b
κ	conductivity ratio, k_o/k_m
λ_n	eigenvalue
σ	standard deviation of asperity heights, μm
ϕ	thermal parameter $\psi_\delta/(\kappa\psi)$
ψ_δ, ψ	thermal constriction parameter, with and without layer

Subscripts

1,2	upper and lower specimen
o,m	oxide and metal

INTRODUCTION

For the most severe loss-of-coolant accident (LOCA) in CANDU reactors, it is predicted that decay heat generated in some of the fuel channels is initially transferred to the cool heavy-water moderator surrounding the fuel channels (1). A key factor in determining the rate of heat transfer is the magnitude of the thermal contact conductance (TCC), or resistance to heat flow, at the interface between the pressure and calandria tubes.

Correlations have been developed (2,3) to predict the steady-state TCC when two clean, conforming, rough surfaces contact under a normal mechanical load. The predictions of these correlations are unreliable, however, when coatings or films are present on the surfaces. In the case of Zr-2.5 wt% Nb pressure tubes, an oxide layer 1-2 μm thick is formed on the outer surface during autoclaving. The thickness of this layer may increase during subsequent normal operation and perhaps during the high-temperature transient associated with certain types of LOCA. Since it is generally known that zirconium oxide is a poor thermal conductor, and much less ductile than the parent metal, its presence on the pressure tube surface would lead to a higher resistance to heat flow, i.e. a lower TCC. This introduces a large uncertainty in our ability to predict TCC with a surface oxide layer present at the pressure tube-calandria tube (PT-CT) interface.

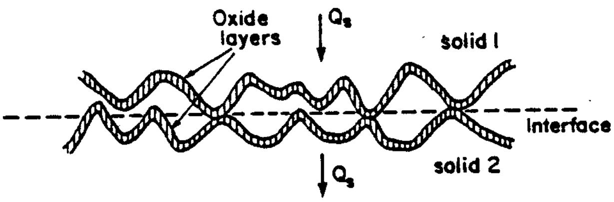
Previous attempts by other investigators (4,5) to account for the influence of oxide layers have met with moderate success in predicting the results of their laboratory experiments. However, their models are cumbersome and tend to mask the physics of the phenomenon, making direct application to the PT-CT problem unfeasible. We desired a more direct approach, in which the role of the important geometric and thermophysical parameters could be clearly defined.

MODELLING AND ANALYSIS

Thermal Model

We consider the problem of determining the TCC for two conforming, rough, oxidized surfaces brought into contact under a normal mechanical load, as shown in Figure 1(a). Out-of-flatness or waviness is ignored, and consideration is given only to heat conduction at the points of contact. A very low pressure or vacuum environment is assumed so that heat transfer through the interstitial gaps is negligible. Radiative heat transfer across the interface is also ignored. It is assumed that the oxide layer is in intimate thermal contact with the substrate and the surface roughness heights can be characterized by a Gaussian distribution: a good assumption for surfaces generated by random processes such as grit blasting or grinding and lapping.

Figure 1(a). Schematic View of Contact Between Nominally Flat, Rough and Oxidized Surfaces



For these conditions, the sole resistance to heat flow at the interface is the constriction of the heat flow lines at each of the points of solid contact, and it is defined as

$$R_\delta = \frac{\Delta T}{Q_s} \tag{1}$$

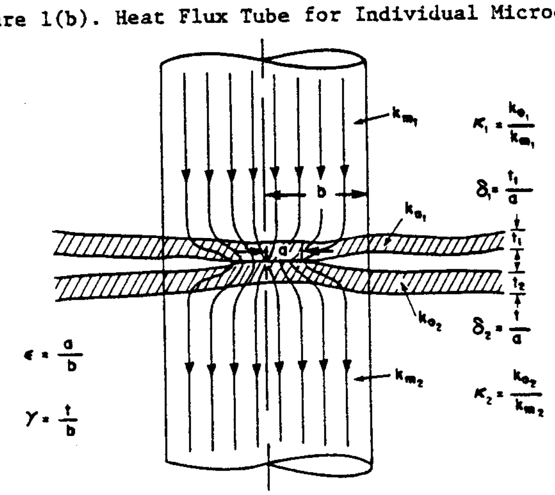
where ΔT is the temperature drop required to overcome the thermal resistance of the contacts and Q_s is the steady-state heat flow. The TCC is defined as

$$h_\delta = \frac{(Q_s/A_a)}{\Delta T} = \frac{1}{R_\delta A_a} \tag{2}$$

where A_a is the total projected area of the surface perpendicular to the direction of flow - commonly referred to as the apparent area of contact.

This complex thermal problem is made tractable by visualizing the total heat flow separated into individual flows corresponding to each microcontact and its associated gap. We can then define an elemental heat flux tube, of radius b, as a volume that encompasses a microcontact, of radius a, and extends some distance into both solids, as indicated in Figure 1(b). The problem is thus reduced to heat flow through a single circular contact-spot located centrally in a compound cylindrical region. Application of this thermal model to a number of microcontact spots appropriately distributed over an area of apparent contact, A_a, between real solids will be discussed in a later section.

Figure 1(b). Heat Flux Tube for Individual Microcontact



A general expression for determining the thermal constriction resistance of a circular area supplying heat to a compound right-circular cylinder has been obtained by Yovanovich, Tien and Schneider (6). From this work the constriction resistance of one-half of the cylinder, composed of a hard resistive layer (oxide) on a metal substrate, is given by

$$R_\delta = \frac{2}{a k_o \pi \epsilon} \sum_{n=1}^{\infty} \left[\frac{\exp(2\lambda_n \gamma) + \beta}{\exp(2\lambda_n \gamma) - \beta}\right] \frac{J_1(\lambda_n \epsilon)\sin(\lambda_n \epsilon)}{\lambda_n^3 J_o^2(\lambda_n)} \tag{3}$$

where $\beta = (\kappa-1)/(\kappa+1)$ and $\kappa = k_o/k_m$. This expression has been derived assuming an infinitely long cylinder with an isothermal temperature prescribed over the circular contact. The remainder of the cylinder surface is assumed to be impervious to heat transfer.

A non-dimensional constriction parameter is defined as $\psi_\delta = 4ak_oR_\delta$, so that the contact conductance for one-half of the flux tube shown in Figure 1(b) is given by

$$h_\delta = \frac{4ak_o}{\pi b^2}\frac{1}{\psi_\delta} \tag{4}$$

Values of the constriction parameter, ψ_δ, for the range of geometric and thermal parameters of interest in this investigation were obtained by programming the expression given in equation (3) for summation on a CDC-6600 computer. Some results are shown in Figure 2, where ψ_δ is plotted as a function of the ratio of the layer thickness to the contact-spot radius, $\delta = t/a$, for conductivity ratios of $\kappa = 0.01$ and $\kappa = 0.1$. The effect of varying the ratio of contact-spot radius to flux-tube radius, $\epsilon = a/b$, is also shown. We see that the constriction parameter increases (h_δ decreases) with increasing δ and approaches unity when $\delta \geq 10$. In fact, when $\delta \geq 2.0$, ψ_δ has achieved about 80% of its maximum value, indicating that most of the constriction is occurring in the oxide layer and the contact conductance is approaching the value $h_\delta = 4ak_o/\pi b^2$.

Figure 2. Constriction Parameter vs Relative Layer Thickness

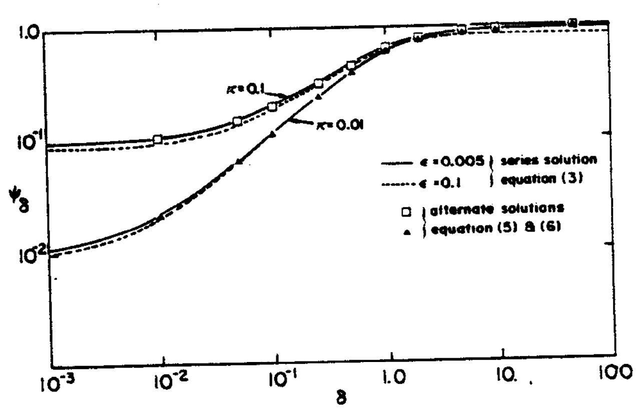
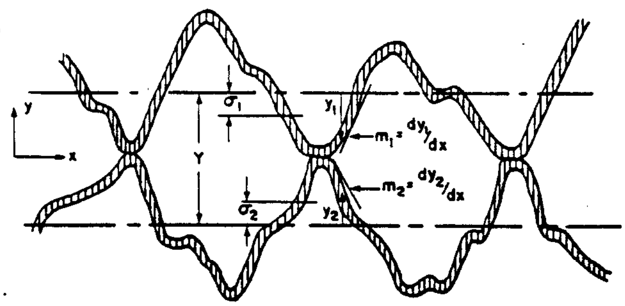


Figure 3. Profiles of Contacting Surfaces (Vertical Scale Magnified)



These relationships are used to obtain an expression for the contact conductance between oxidized surfaces in vacuum, given by

$$h_\delta = \frac{1}{\sqrt{2\pi}} \frac{m}{\sigma} \frac{\exp(-x^2)}{\psi} \cdot \left[\frac{1}{\frac{\Phi_1}{k_{m_1}} + \frac{\Phi_2}{k_{m_2}}} \right] \quad (13)$$

where $x = \text{erfc}^{-1}(P_a/H_e)$ and $\varepsilon = (P_a/H_e)^{1/2}$

APPLICATION OF LAYER MODEL

To simplify application of this model, we will consider a special case in which both solids are the same material and only the lower specimen surface is covered with an oxide layer. In this case $\delta_1 = 0$, $\Phi_1 =$ 1 and $k_{m_1} = k_{m_2}$. Then equation (13) can be written in non-dimensional form as

$$\frac{\sigma h_\delta}{m k_s} = \left(\frac{1}{2\sqrt{2\pi}} \frac{\exp(-x^2)}{\psi} \right) \left[\frac{2}{(1 + \Phi_2)} \right] \quad (14)$$

where $k_s = \frac{2k_{m_1}k_{m_2}}{k_{m_1}+k_{m_2}}$ is the harmonic mean thermal conductivity of the substrates.

Referring to earlier work by Yovanovich (3), the term in round brackets can be expressed in terms of P_a/H_e by the simple correlation 1.25 $(P_a/H_e)^{0.95}$, so that equation (14) becomes

$$\frac{\sigma h_\delta}{m k_s} = 1.25 \left(\frac{P_a}{H_e} \right)^{0.95} \left[\frac{2}{(1 + \Phi_2)} \right] \quad (15)$$

We see that the term $2/(1 + \Phi_2)$ modifies the correlation for conforming rough surfaces to account for the effect of the layer. This effect is dominated by the magnitude of the thermal conductivity ratio, but there is some dependence on P_a/H_e through the ratio ψ_δ/ψ.

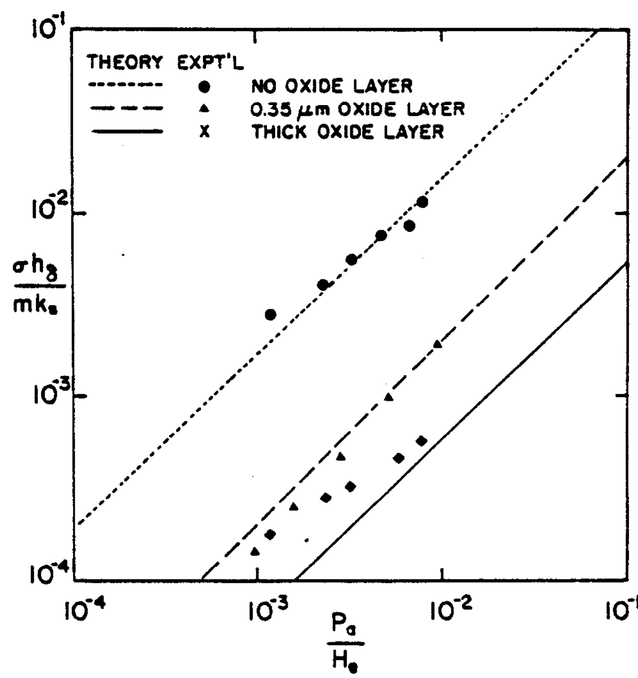
We compared this correlation with results reported in the literature by Mian et al (9) for oxidized steel surfaces. Pertinent geometric and thermal parameters are given in Table 1. For the effective hardness, H, we used the value for steel, since it is softer than the oxidized surface. The reported value was increased to reflect the surface hardening effect observed by Yovanovich, Hegazy and Devaal (10) for metal surfaces prepared by grinding and lapping.

Table 1. Geometric and Thermal Parameters of Samples From Reference 9

Specimen Pair	σ (μm)	m	H_e (MPa)	t (μm)	k_m W/(m.°C)	k_o W/(m.°C)
a	0.629	0.059	2610	-	47.0	-
b	0.144	0.046	2610	0.35	47.0	0.875
c	2.326	0.214	2610	very thick	47.0	0.875

Comparisons were made with measurements from specimens with (a) no oxide layer, (b) a thin 0.35 μm layer and (c) a very thick layer for which it is assumed that all of the constriction occurred in the oxide layer. The good agreement with the predictions of equation (15), shown in Figure 4, is reassuring and indicates that the model has successfully incorporated the effects of the oxide layer. The poorer agreement with the very thick layer was not unexpected since Mian et al (9) reported that the surface roughness deviated from a Gaussian distribution.

Figure 4. Comparison of Experimental and Theoretical Results of TCC for an Oxidized Steel Surface



TCC BETWEEN OXIDIZED ZR-2.5 WT% NB AND ZIRCALOY-4

Experimental Measurements

Cylindrical specimens of Zr-2.5 wt% Nb and Zr-4, 25.4 mm in diameter and 25.4 mm long, were cut from bar stock, and the ends were ground and lapped to produce nominally flat, smooth surfaces.

The Zr-2.5 wt% Nb specimens were prepared for controlled oxide growth in a high-temperature autoclave by ultrasonic cleaning in acetone, followed by isopropyl alcohol. The surfaces were then swabbed with a caustic wash to remove any remaining contaminants.

Table 3. Thermophysical Properties for Oxidized Zr-2.5 wt% Nb/Zr-4 Pairs

Specimen	k (W/(m.°C)	H_e (MPa)
Zr-4	15.42	1950
Zr-2.5 wt% Nb	19.54	1607
ZrO_2	4.0	7200

The results show that the conductance decreases with an increase in oxide thickness and that this decrease is controlled by the thermal conductivity of the oxide and the ratio of layer thickness to contact-spot radius. The relative decrease in conductance, compared to the specimen pair without an oxide layer, is practically independent of contact pressure when P_a/H_e is greater than 10^{-3}. These trends agree with the results for oxidized steel pairs reported by Mian et al (9).

CONCLUSIONS

A theoretical model has been developed that adequately describes the effect of oxide layers on the TCC between contacting zirconium alloy surfaces. For the limited range of parameters investigated in this work, this effect appears to simply modify the constant term in the standard correlation for conforming rough surfaces. A thorough investigation over a wider range of geometric, material and thermal parameters is required to confirm this observation and develop a suitable correlation when oxide layers are present at the interface. Further work is also required to reduce the large uncertainty in the value of the thermal conductivity of zirconium oxide, since it is a key parameter in modelling this phenomenon.

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REFERENCES

1. D.A. Meneley and W.T. Hancox, "LOCA Consequence Predictions in a CANDU-PHWR," Report IAEA-CN-42/145, presented at the IAEA International Conference on Nuclear Power Experience, Vienna, September 1982.

2. M.G. Cooper, B.B. Mikic and M.M. Yovanovich, "Thermal Contact Conductance," Int. J. Heat Mass Transfer Vol. 12, pp. 279-300 (1969).

3. M.M. Yovanovich, "New Contact and Gap Conductance Correlations for Conforming Rough Surfaces," presented at American Inst. of Aeronautics and Astronautics 16th Thermophysics Conference, AIAA-81-1164 (1981).

4. F.C. Yip, "The Effect of Oxide Films on Thermal Contact Resistance," AIAA Paper No. 74-693 presented at the AIAA/ASME 1974 Thermophysics and Heat Transfer Conference, pp. 45-64 (1974).

5. F.R. Al-Astrabadi, P.W. O'Callaghan and S.D. Probert, "Thermal Resistance of Contacts: Influence of Oxide Films," presented at American Inst. of Aeronautics and Astronautics 15th Thermophysics Conference, AIAA-80-1467 (1980).

6. M.M. Yovanovich, C.H. Tien and G.E. Schneider, "General Solution of Constriction Resistance Within a Compound Disk," presented at the 17th Aerospace Sciences Meeting, 79-0178 (1979).

7. J. Dryden, personal communication.

8. B. Mikic and G. Carnasciali, "The Effect of Thermal Conductivity of Plating Material on Thermal Contact Resistance," Journal of Heat Transfer, Trans. of the ASME, Vol. 69, pp. 475-482 (1970).

9. M.N. Mian, F.R. Al-Astrabadi, P.W. O'Callaghan and S.D. Probert, "Thermal Resistance of Pressed Contacts Between Steel Surfaces: Influence of Oxide Films," Journal of Mechanical Engineering Science, Vol. 21, No. 3, pp. 159-166 (1979).

10. M.M. Yovanovich, A.H. Hegazy and J. DeVaal, "Surface Hardness Distribution Effects Upon Contact, Gap and Joint Conductances," presented at AIAA/ASME 3rd Joint Thermophysics, Fluids, Plasma and Heat Transfer Conference, Paper No. AIAA-82-0887 (1982).

11. K.E. Gilchrist, "Thermal Property Measurements on Zircaloy-2 and Associated Oxide Layers up to 1200°C," Journal of Nuclear Materials, Vol. 62, pp. 257-264 (1976).

12. H. Maki, "Heat Transfer Characteristics of Zircaloy-2 Oxide Film," Journal of Nuclear Science and Technology, Vol. 10, No. 3, pp. 170-175 (1973).