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**TRANSIENT ENERGY TRANSFER BY CONDUCTION AND  
RADIATION FOR A SUDDEN CONTACT BETWEEN  
MOLTEN UO<sub>2</sub> AND SODIUM**

by

**D. H. Cho and S. H. Chan**

BASE TECHNOLOGY

MASTER



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**ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS**

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D. H. Cho and S. H. Chan\*

Reactor Analysis and Safety Division

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NOMENCLATURE

B	interface radiosity
c	specific heat
$E_n$	exponential integral of the nth order
H	incident radiation on the interface
k	radiative absorption coefficient
n	refractive index
p	Laplace-transform variable
q	heat flux
$q_c$	conductive heat flux
$q_r$	radiative heat flux
T	temperature
t	time
$t^*$	dimensionless time
x	distance
$\delta_1, \delta_2$	dimensionless parameters
$\epsilon$	interface emissivity
$\theta$	dimensionless temperature
$\theta_r$	dimensionless temperature representing the interface radiosity effect
$\lambda$	thermal conductivity
$\xi$	optical depth
$\rho$	density
$\rho_r$	interface reflectivity
$\sigma$	Stefan-Boltzmann constant
$\tau$	modified optical depth
$\phi_1, \phi_2$	dimensionless parameters
Subscripts	
c	cold medium
h	hot medium
i	interface
o	initial value

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ABSTRACT

The transient energy transfer following a sudden contact between molten  $UO_2$  and sodium has been investigated, taking into consideration both conduction and internal thermal radiation in  $UO_2$ . Analytical expressions for the contact-interface temperature valid for small times are derived. Illustrative calculations indicate that on a time scale relevant to fuel-coolant interactions, internal radiation of molten  $UO_2$  should have an insignificant effect on the contact-interface temperature between molten  $UO_2$  and sodium. It thus appears that for the purpose of assessing the potential for an explosive fuel-coolant interaction, the contact-interface temperature may be adequately determined based on consideration of pure conduction.

## I. INTRODUCTION

The interface temperature that results when two media at different temperatures are suddenly brought into contact is an important consideration in evaluating the potential for explosive boiling in liquid-liquid systems.<sup>1</sup> As such, it is of interest in safety studies of liquid-metal-cooled fast breeder reactors since molten reactor fuel may come into contact with the sodium coolant in accident situations. The contact-interface temperature also plays a role in the development of stable film boiling.<sup>2</sup> Usually the Fourier conduction equation is employed to determine the contact-interface temperature. Recently, Kim *et al.*<sup>3</sup> measured the thermal diffusivity of molten  $UO_2$  by the modulated electron-beam technique. The thermal diffusivity was found to be about  $2.40 \times 10^6 \text{ m}^2/\text{s}$  in the liquid region near the melting point. This value is much larger than the thermal diffusivities of  $UO_2$  reported in the solid region at lower temperatures ( $\sim 0.5 \times 10^6 \text{ m}^2/\text{s}$ ). The high value of thermal diffusivity for molten  $UO_2$  was attributed to the contribution of internal radiation to heat transfer in molten  $UO_2$ . The purpose of this report is to examine the effect of this additional mode of heat transfer on the contact-interface temperature. Internal radiation in the cold medium is not considered. Analytical solutions valid for small times are derived using an approximate method due to Lick.<sup>4</sup> In this method, the radiative-flux term is linearized and the basic integro-differential equation governing the energy transfer by radiation and conduction is converted to a differential equation by a degenerate-kernel approximation technique. Illustrative calculations are presented for a sudden contact between molten uranium dioxide and sodium. The calculations indicate that, on a time scale relevant to fuel-coolant interactions, internal radiation of molten  $UO_2$  should have insignificant effect on the contact-interface temperature.

## II. GOVERNING EQUATIONS

Consider a semi-infinite medium capable of transferring energy by radiation and conduction. Initially the medium is at a uniform temperature  $T_{ho}$  throughout. At time  $t = 0$ , the medium is suddenly brought into contact with another medium at a uniform temperature  $T_{co}$  which is lower than  $T_{ho}$ . In the cold medium, conduction is considered to be the only mechanism for energy transfer.

The governing equation for the hot medium is

$$-\frac{\partial q}{\partial x} = \rho_h c_h \frac{\partial T_h}{\partial t}, \quad (1)$$

where the energy flux  $q$  is the sum of the conductive heat flux  $q_c$  and the radiative heat flux  $q_r$ . The conductive heat flux is given by

$$q_c = -\lambda_h \frac{\partial T_h}{\partial x} \quad (2)$$

and the radiative heat flux by

$$q_r = 2n^2\sigma \int_0^\xi T_h^4(\bar{\xi}) E_2(\xi - \bar{\xi}) d\bar{\xi} - 2n^2\sigma \int_\xi^\infty T_h^4(\bar{\xi}) E_2(\bar{\xi} - \xi) d\bar{\xi} + 2B E_3(\xi) \quad (3)$$

where  $E_n$  is the exponential integral

$$E_n(\xi) = \int_0^1 \mu^{n-2} \exp(-\xi/\mu) d\mu \quad (4)$$

and  $\xi$  is the optical depth:  $\xi = kx$ . It has been assumed in writing equation (3) that the absorption coefficient  $k$  is independent of frequency and temperature. The radiosity of the contact interface is denoted by  $B$ . If it is assumed that the interface is gray, and that the radiation reflected and emitted from the interface is diffusely distributed, the radiosity is given by

$$B = \epsilon n^2 \sigma T_i^4 + \rho_r H, \quad (5)$$

where  $\epsilon$  and  $\rho_r$  are the emissivity and the reflectivity of the interface,

respectively. The incident radiation arriving at the interface from the interior of the hot medium is given by

$$H = 2n^2\sigma \int_0^\infty T_h^4 E_2(\xi) d\xi , \quad (6)$$

In writing equation (5) it has been implicitly assumed that the cold medium is opaque, so only surface radiation is considered.

The energy transfer in the cold medium is governed by the usual Fourier conduction equation:

$$\lambda_c \frac{\partial^2 T_c}{\partial x^2} = \rho_c c_c \frac{\partial T_c}{\partial t} , \quad (7)$$

Following Lick's approach,<sup>4</sup> we linearize the radiative-flux term and make a kernel substitution. An approximate kernel of the form  $(3/4)\exp[-(3/2)x]$  is substituted for  $E_2(x)$  and  $(1/2)\exp[-(3/2)x]$  for  $E_3(x)$ . The resulting governing equation for the hot medium then becomes, in dimensionless form,

$$\delta_1 \frac{\partial^2 \theta_h}{\partial \tau^2} - \frac{\partial \theta_h}{\partial \tau^*} = -2 \int_0^\tau \theta_h \exp[-(\tau - \bar{\tau})] d\bar{\tau} - 2 \int_\tau^\infty \theta_h \exp[-(\bar{\tau} - \tau)] d\bar{\tau} + 4\theta_h - 2\theta_r \exp(-\tau) , \quad (8)$$

where  $\theta_h = (T_h/T_{ho}) - 1$ ,  $\tau = (3/2)\xi$ , and  $\tau^* = \frac{3kn^2\sigma T_{ho}^3 t}{\rho_h c_h}$ .

The dimensionless parameter

$$\delta_1 = \frac{3\lambda_h k}{4n^2\sigma T_{ho}^3} \quad (9)$$

characterizes the ratio of the amount of heat conducted and the amount of heat radiated. The dimensionless temperature  $\theta_r$  is given by

$$\theta_r = \frac{1}{4} \left( \frac{B}{n^2\sigma T_{ho}^4} - 1 \right) \quad (10)$$

and represents the effect of the radiosity of the interface. It may be noted that the radiosity  $B$  is a function of time inasmuch as the interface temperature  $T_i$  and the incident radiation  $H$  vary with time.

As was done by Lick, we differentiate equation (8) twice with respect to  $\tau$  and substitute the resulting equation back into equation (8) to eliminate the integral terms. We then obtain

$$\delta_1 \frac{\partial^4 \theta_h}{\partial \tau^4} - (\delta_1 + 4) \frac{\partial^2 \theta_h}{\partial \tau^2} = \frac{\partial^3 \theta_h}{\partial \tau^2 \partial \tau^*} - \frac{\partial \theta_h}{\partial \tau^*}, \quad (11)$$

The conduction equation for the cold medium becomes, in dimensionless form,

$$\frac{\partial \theta_c}{\partial \tau^*} = \delta_2 \frac{\partial^2 \theta_c}{\partial \tau^2}, \quad (12)$$

where

$$\theta_c = \frac{T_c}{T_{ho}} - \frac{T_{co}}{T_{ho}}; \quad \delta_2 = \left( \frac{\lambda_c \rho_h c_h}{\lambda_h \rho_c c_c} \right) \delta_1.$$

The boundary conditions appropriate to the problem are

$$\tau^* = 0: \quad \theta_h = \theta_c = 0;$$

$$\tau \rightarrow \infty: \quad \theta_h \rightarrow 0;$$

$$\tau = 0: \quad \theta_h + 1 = \theta_c + \frac{T_{co}}{T_{ho}};$$

$$\frac{\lambda_c}{\lambda_h} \frac{\partial \theta_c}{\partial \tau} = \frac{\partial \theta_h}{\partial \tau} + \frac{1}{2\delta_1} \left[ \frac{H-B}{n^2 \sigma T_{ho}^4} \right];$$

$$\tau \rightarrow -\infty: \quad \theta_c \rightarrow 0.$$

The second condition for  $\tau = 0$  represents continuity in the heat flux at the interface.

The differential equations (11) and (12) may be solved by the Laplace-transform method. As shown by Lick the solution of the Laplace-transform of equation (11) is

$$\bar{\theta}_h = A_1 \exp(\gamma_1 \tau) + A_2 \exp(\gamma_2 \tau). \quad (13)$$

For small times the two roots  $\gamma_1$  and  $\gamma_2$  may be approximated by

$$\gamma_1 = - \left( \frac{p}{\delta_1} \right)^{1/2} - \frac{2}{\delta_1^{1/2} p^{1/2}} + 0 \left( \frac{1}{p^{3/2}} \right) \quad (14)$$

and

$$\gamma_2 = - 1 + \frac{2}{p} + 0 \left( \frac{1}{p^2} \right), \quad (15)$$

where  $p$  is the Laplace-transform variable. A relationship between the integration constants  $A_1$  and  $A_2$  can be found by requiring that the solution, equation (13), satisfy the Laplace transform of the integro-differential equation (8). The relationship is

$$\frac{A_1}{\gamma_1 + 1} + \frac{A_2}{\gamma_2 + 1} - L \left\{ \theta_r \right\} = 0, \quad (16)$$

where  $L \left\{ \theta_r \right\}$  is the Laplace transform of  $\theta_r$ .

The solution of the Laplace transform of equation (12) is

$$\bar{\theta}_c = B_1 \exp \left( \sqrt{\frac{p}{\delta_2}} \tau \right) \quad (17)$$

The integration constant  $B_1$  is related to  $A_1$  and  $A_2$  via the interface matching conditions. If we apply a Laplace transformation to the boundary conditions at  $\tau = 0$ , and substitute equations (13) and (17) into the transformed boundary conditions, we obtain

$$A_1 + A_2 + \frac{1}{p} = B_1 + \left( \frac{T_{co}}{T_{ho}} \right) \frac{1}{p} \quad (18)$$

and

$$\frac{\lambda_c}{\lambda_h} B_1 \sqrt{\frac{p}{\delta_2}} = A_1 \gamma_1 + A_2 \gamma_2 + \frac{1}{2\delta_1} \frac{L \left\{ H-B \right\}}{n^2 \sigma T_{ho}^4}. \quad (19)$$

The constants  $A_1$ ,  $A_2$ , and  $B_1$  may, in principle, be evaluated from equations (16), (18), and (19). However, this cannot be done explicitly since  $H$  and  $B$  involve the interface temperature as well as the temperature distribution in the hot medium and thus are unknown. In the following we shall consider two simplifications.

### III. SOLUTION FOR A COMPLETE REFLECTION AT THE INTERFACE

The incident radiation coming from the interior of the hot medium is considered to be completely reflected back from the interface, i.e.,  $\rho_r = 1$ . It then follows from the gray-body assumption that  $\epsilon = 0$ . In this case, the radiosity  $B$  becomes

$$B = H = 2n^2\sigma \int_0^{\infty} T_h^4 E_2(\xi) d\xi. \quad (20)$$

If we further approximate that

$$\int_0^{\infty} T_h^4 E_2(\xi) d\xi \approx T_{ho}^4 \int_0^{\infty} E_2(\xi) d\xi = \frac{T_{ho}^4}{2}, \quad (21)^*$$

we find  $\theta_r = 0$ . For  $\theta_r = 0$ , equation (16) indicates that

$$\frac{A_2}{A_1} = 0 \left( \frac{1}{p^{3/2}} \right)$$

for small times, so  $A_2$  may be ignored compared to  $A_1$ . With this approximation constants  $A_1$  and  $B_1$  are readily obtained from equations (18) and (19). For small times,

$$A_1 = \frac{-\phi_2 (1 - T_{co}/T_{ho})}{p + 2\phi_1} \quad (22)$$

and

$$B_1 = \left( 1 - \frac{T_{co}}{T_{ho}} \right) \left( \frac{1}{p} - \frac{\phi_2}{p + 2\phi_1} \right), \quad (23)$$

where  $\phi_1 = \frac{\sqrt{\lambda_h \rho_h c_h}}{\sqrt{\lambda_h \rho_h c_h} + \sqrt{\lambda_c \rho_c c_c}}$  and  $\phi_2 = 1 - \phi_1$ .

---

\*This approximation is probably reasonable for small times.

The expressions for  $A_1$  and  $B_1$  are substituted into equations (13) and (17), and an inverse transformation of the resulting equations gives the temperature profiles in the two media. The interface temperature  $T_i$  is given by

$$\frac{T_i - T_{ho}}{(T_i)_c - T_{ho}} = \exp(-2\phi_1 t^*) , \quad (24)$$

where  $(T_i)_c$  is the interface temperature based on the Fourier conduction solution. The temperature profiles are approximately given by, for  $\tau > 0$ ,

$$\begin{aligned} \frac{T_h - T_{ho}}{(T_i)_c - T_{ho}} &= \operatorname{erfc} \left( \frac{\tau}{2\sqrt{\delta_1 t^*}} \right) - (4t^*)^{1/2} \frac{2\tau}{\delta_1^{1/2}} i \operatorname{erfc} \left( \frac{\tau}{2\sqrt{\delta_1 t^*}} \right) \\ &- 2\phi_1 \left[ (t^* + \frac{\tau^2}{2\delta_1}) \operatorname{erfc} \left( \frac{\tau}{2\sqrt{\delta_1 t^*}} \right) - \tau \left( \frac{t^*}{\pi\delta_1} \right)^{1/2} \exp \left( -\frac{\tau^2}{4\delta_1 t^*} \right) \right. \\ &\left. - (4t^*)^{3/2} \left( \frac{2\tau}{\delta_1^{1/2}} \right) i^3 \operatorname{erfc} \left( \frac{\tau}{2\sqrt{\delta_1 t^*}} \right) \right] , \end{aligned} \quad (25)$$

and, for  $\tau < 0$ ,

$$\begin{aligned} \frac{T_c - T_{co}}{(T_i)_c - T_{co}} &= \operatorname{erfc} \left( \frac{-\tau}{2\sqrt{\delta_2 t^*}} \right) + 2\phi_2 \left[ (t^* + \frac{\tau^2}{2\delta_2}) \operatorname{erfc} \left( \frac{-\tau}{2\sqrt{\delta_2 t^*}} \right) \right. \\ &\left. + \tau \left( \frac{t^*}{\pi\delta_2} \right)^{1/2} \exp \left( -\frac{\tau^2}{4\delta_2 t^*} \right) \right] . \end{aligned} \quad (26)$$

In deriving these solutions, certain linearizations of equations (13) and (17) have been made. It may be noted that the first two terms of equation (25) correspond to Lick's solution<sup>4</sup> for a constant surface temperature (i.e.,  $\phi_1 = 0$  and  $(T_i)_c = T_{co}$ ).

#### IV. SOLUTION FOR A PARTIAL REFLECTION AT THE INTERFACE

We now consider the case when the incident radiation arriving at the interface is partially reflected back into the hot medium. We shall make the approximation  $H = n^2 \sigma T_{ho}^4$  as before, and ignore the emission from the interface compared to the incident radiation. This assumption would be reasonable when the interface temperature is much lower than the bulk temperature of the hot medium. We then find  $\theta_r = (\rho_r - 1)/4$ . Proceeding as before and ignoring higher order terms than  $1/p^2$  in the expressions for  $A_1$  and  $A_2$ , we obtain

$$A_1 = \frac{-\phi_2(1-T_{co}/T_{ho}) + \phi_1(1-\rho_r)/(2\sqrt{\delta_1 p}) + \phi_2(1-\rho_r)/(2p)}{p + 2\phi_1} \quad (27)$$

and

$$A_2 = -\frac{(1-\rho_r)}{2p^2} \quad (28)$$

The interface temperature is obtained by inverting the expression for  $(A_1 + A_2)$  and is given by

$$\begin{aligned} T_i - T_{ho} &= [(T_i)_c - T_{ho}] \exp(-2\phi_1 t^*) \\ &+ T_{ho}(1-\rho_r) \left[ \frac{\phi_1}{\sqrt{2\pi\delta_1\phi_1}} \exp(-2\phi_1 t^*) \int_0^{\sqrt{2\phi_1 t^*}} \exp(\lambda^2) d\lambda \right. \\ &\left. + \frac{1}{4} \frac{\phi_2}{\phi_1} [1 - \exp(-2\phi_1 t^*)] - \frac{1}{2} t^* \right] \end{aligned} \quad (29)$$

For  $\rho_r = 1$  equation (29) reduces to equation (24).

## V. ILLUSTRATIVE CALCULATIONS AND DISCUSSION

Illustrative calculations have been made for a sudden contact between molten uranium dioxide and sodium initially at 3100 K and 1100 K, respectively. The contact-interface temperature based on the Fourier conduction solution is 1710 K, which remains constant for all times. One of the key parameters for the calculations is the radiative absorption coefficient  $k$ . At present no experimental data are available for the optical properties of molten uranium dioxide. It has been suggested that, since the molten state contains no scattering centers such as voids or grain boundaries, the molten-state spectrum should resemble that of a single crystal.<sup>5</sup> Based on this suggestion an estimated value of  $50 \text{ cm}^{-1}$  was used for the absorption coefficient. The refractive index  $n$  of uranium dioxide has been measured by Ackermann *et al.*<sup>6</sup>, and they found values between 2.1 and 2.5. In the present calculations the refractive index was taken to be 2.3.

The calculational results are presented in Fig. 1. Immediately following the contact, the interface temperature is equal to the value based on the Fourier conduction solution. With increasing time, however, internal radiation in the  $\text{UO}_2$  becomes important, and the interface temperature increases above the pure conduction value. The increase 2 ms after the contact is about 50 K for  $\rho_r = 1.0$  and somewhat greater for lower values of  $\rho_r$ . The assumption that the emission from the interface is negligible seems justified for the  $\text{UO}_2$ -sodium system, since the interface temperature is much lower than the bulk temperature of molten  $\text{UO}_2$ . For the present calculations, the maximum amount of emission from the interface for small times is about  $(17/31)^4$  or 9% of the incident radiation.

The time scale for contact relevant to an explosive interaction between molten fuel and coolant is believed to be of the order of 1 ms or less.<sup>1</sup> The above calculations seem to indicate that on this time scale, internal radiation of molten  $\text{UO}_2$  should have insignificant effect on the contact-interface temperature between molten  $\text{UO}_2$  and sodium. The reflectivity of the  $\text{UO}_2$ -sodium interface has been estimated to be 0.965 at 1700 K<sup>7</sup>. By use of this value of  $\rho_r$ , the contact-interface temperature 1 ms after the contact is found to be 1738 K, which is only 28 K above the pure conduction value and

well below the homogeneous nucleation temperature of sodium ( $\sim 2300$  K). It thus appears that for the purpose of assessing the potential for an explosive fuel-coolant interaction, the contact-interface temperature may be adequately determined based on consideration of pure conduction. However, internal radiation in molten fuel might play an important role in certain phase-change and heat-transfer processes relevant to postaccident heat removal in a fast-breeder nuclear reactor, where the time scale is of the order of many seconds. In this case, it would also be necessary to know the reflection, absorption, and emission characteristics of the fuel and its bounding interfaces with other media, such as steel and sodium.<sup>7</sup>

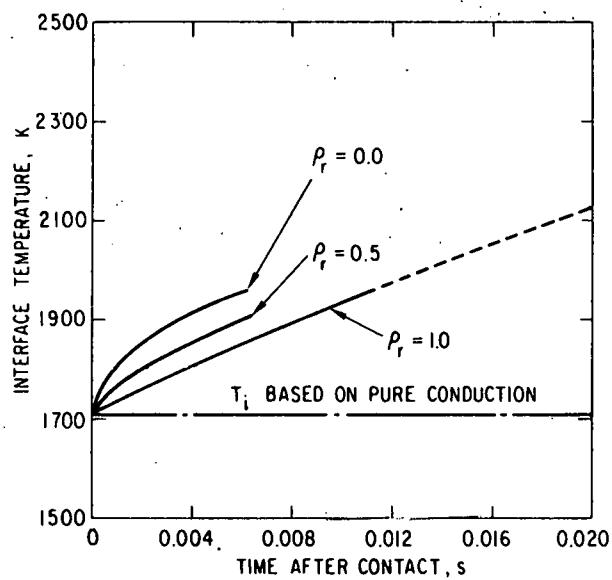


Fig. 1. Effect of Internal Radiation in Molten  $UO_2$  on the Contact-interface Temperature between  $UO_2$  and Sodium. (In the broken portion of the curve for  $\rho_r = 1.0$ , the validity of the solution is questionable.)

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