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AUTHOR(S): Anthony D. Rollett, MST-6 Homer D. Lewis, MST-6 Paul S. Dunn, MST-6

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# MEASUREMENT AND MODELING OF INTERFACE HEAT TRANSFER COEFFICIENTS

A. D. Rollett, H. D. Lewis and P. S. Dunn

# Materials Science and Technology Division Los Alamos National Laboratory Los Alamos, NM 87545

#### Summary

The results of preliminary work on the modeling and measurement of the heat transfer coefficients of metal/mold interfaces is reported. The system investigated is the casting of uranium in graphite molds. The motivation for the work is primarily to improve the accuracy of process modeling of prototype mold designs at the Los Alamos Foundry. The evolution in design of a suitable mold for unidirectional solidification is described, illustrating the value of simulating mold designs prior to use. Experiment indicated a heat transfer coefficient of 2 kW/m<sup>2</sup>/K both with and without superheat. It was possible to distinguish between solidification fication due to the mold and that due to radiative heat loss. This perimited an experimental estimate of the emissivity,  $\varepsilon = 0.2$ , of the solidified metal.

#### Introduction

A feature that distinguishes casting processes from welding processes is the presence of an interface with a low heat transfer coefficient between the mold and the casting. Especially when chill casting, the interface delays the start of solidification and increases freezing times signifi-By comparison welds can be assumed to have a very high heat cantly. transfer coefficient. The heat transfer coefficient controls such phenomena as folds and laps on the surface of castings which can be crucial to the acceptability of a casting. The time delay for the start of solidification influences the occurrence of cold shut defects. The standard mold material for uranium casting is high quality graphite which is an excellent refractory when used in vacuum and coated with a refractory Graphite is an effective chill material and has a thermal wash. diffusivity approximately twice that of uranium. Experience has shown that practicable superheat without mold pre-heat is not sufficient for avoiding cold shut defects when casting in graphite molds.

#### Theory

It is possible to calculate the position of the freeze front s, as a function of time, t, the superheat and interface heat transfer coefficient, h, [1]. However, the value of h cannot be obtained explicitly. The Virtual Adjunct Method (VAM) [2] offers a method for extending the standard equation for an insulating mold where s is proportional to the square root of time. By considering the effect of the interface to be equivalent to finite thicknesses of mold and metal, the following quadratic equation is obtained,

$$t = As^2 + Bs.$$
(1)

The constants A and B are given by,

$$1/A = 4a_{s}\phi^{2}$$
 (2)

$$B = Ld_{s}/((T_{f}-T_{o})h).$$
 (3)

T, is the freezing temperature of the metal and To is the initial temperature of the mold.

L is the latent heat of fusion of the metal and  $\phi$  is given by the standard expression for a semi-infinite mold, ref. 3, equation 9b, as,

$$(\pi)^{1/2} \phi \exp(\phi^2) [erf(\phi) + M] = c_s (T_f - T_o) / L,$$
 (4)

where

$$M = (k_{s} d_{s} c_{s}/k_{m} d_{m} c_{m})^{1/2}.$$
 (5)

The variables k, d, c and a are thermal conductivity, density, specific heat and thermal diffusivity, respectively. Subscripts s and m denote properties for metal and mold, respectively. The necessary requirement for equation (1) to be valid is that there be no superheat present in the liquid metal. The presence of superheat leads to a delay in the start of freezing due to the large effective thermal conductivity of the convecting melt. Convection in the liquid metal also tends to dissipate the superheat before a significant amount of solidification occurs.

Clemente and Vicente [4] have criticized the VAM because the method incorrectly assumes that the interface is equivalent to a fixed, virtual amount of mold and metal thickness, S. The point can be demonstrated qualitatively as follows; for a constant heat transfer coefficient, a fixed imaginary thickness of mold plus metal means that the temperature drop across the interface must remain constant. However, at large times the temperature at the metal side of the interface will be falling. In the case of the cooled mold where the temperature of the mold side remains approximately constant this implies that the temperature differential across the interface decreases. Hence the virtual thickness, S., also decreases. This can be shown by writing a heat balance at the interface.

For simplicity, consider the case of the cooled mold. Following the equations of reference [1], the temperature of the metal side of the interface,  $T_4$ , is,

$$T_{i} = T_{o} + (T_{f} - T_{o}) \operatorname{erf}(\phi S_{o} / (s + S_{o})) / \operatorname{erf}(\phi)$$
(6)

where S is the "virtual adjunct" of effective pre-existing solid metal thickness. The thermal gradient at that point, dT/dx, is

$$dT/dx = 2\phi (T_f - T_o) / [(\pi)^{1/2} S_o erf(\phi) exp(\phi^2)].$$
 (7)

The heat flux crossing the interface is h  $(T_i-T_i)$ . The flux must be equal to the heat supplied by the solid metal,  $k_s$  dT/dx, at the interface. This leads to an equation of the form,

$$k_{s} 2\phi/[h(\pi)^{1/2} exp(\phi^{2})] = [S_{o}+s] erf(\phi S_{o}/[S_{o}+s]) (8)$$

The thermal properties of the mold/metal system fix  $\bullet$  and h is presumed to be constant. Therefore S must vary (decrease) as the solidified thickness, s, increases with time. Given that the interface conductance remains constant, this implies that the temperature difference across the interface decreases. This is consistent with experimental observations. The freeze front position, s, calculated by equation 1 is not an explicit function of S. The theory gave excellent agreement with the experimental results of Garcia et. al. [3]. An assumption of the VAM is not correct, as pointed out by Clemente & Vicente. However the VAM does provide a useful analytical method for estimating the interfacial heat transfer coefficient. The interesting feature of the Virtual Adjunct Method, for the case of the cooled mold, is that it gives the same time dependence for thickness solidified as the simpler assumption of a linear temperature gradient in the solid metal. The work described here, however, makes use of the solution for the semi-infinite mold.

### Modeling

Solidification modeling has already been performed at Los Alamos with the ABAQUS finite element code [5] and the associated software. The applicability to the experiments reported here was tested by simulating the experiments of Garcia et. al. The results are displayed in Fig. 1 which shows both the theoretical lines, the experimental data and the simulated freezing times calculated by ABAQUS. The time plotted is the time at which the metal temperature cools through the liquidus temperature at the given position.



Figure 1. Freezing time over distance plotted versus distance from interface for Pb solidifying in a steel mold [3]. Circles and squares are experimental points for coated and polished mold surfaces, respectively. Crosses and diamonds are simulated points for the same two conditions. Solid and dotted lines are Virtual Adjunct Method theory for the two conditions. The simulated data for the polished mold with a high heat transfer coefficient gave excellent agreement with the experiment. The simulation of the coated mold was adequate. The simulated freezing times were slightly over estimated. Note that the slope of each line, constant A in equation 1, is determined by the thermal properties of the metal and mold whereas the intercept on the y-axis, constant S in equation 1, is inversely proportional to the interfacial heat transfer coefficient.

Above 1000°C, radiative heat transfer was expected to be a significant fraction of the effective interface heat transfer coefficient. A simulation of uranium freezing in a graphite mold with 8°C superheat is illustrated in Fig. 2.



Figure 2. Freezing time over distance plotted versus distance from interface for uranium solidifying in a graphite mold. Squares are simulated points. Solid line is calculated from VAM theory. The simulated point for the furthest distance from the interface has a short freezing time because of radiative heat loss from the top of the casting.

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The intercept indicates a coefficient of  $500 \text{ W/m}^2/\text{K}$ . However the simulation also included radiation across the gap with an emissivity of 0.9. This should have raised the effective coefficient to 874 W/m<sup>2</sup>/k, suggesting the same discrepancy as observed in the comparison of of simulation and experiment data for a coated mold illustrated in Fig. 1.

The solidification model was used to design the mold. The final refinement of the design was to extend the brick lining down into the mold below the base of the metal cavity. Figures 3 and 4 show isotherms before and after this design modification.



Figure 3. Isotherm plots for a tentative mold design. Isotherms 5 and 6 define a narrow mushy zone. Although the freeze fronts are nearly flat, heat flow is not as unidirectional as desired.

Figure 4. Isotherm plot for the mold design used. Note the improvement in heat flow pattern due to extending the brick insulation downwards. Solidification is nearly complete

The corner effects and the lack of unidirectional solidification evident in Fig. 3 were much improved by the change in mold design as shown in Fig. 4.

#### Experimental

The mold was a cylinder machined from a single piece of graphite. The cavity was lined with a standard refractory brick, heavily coated with zirconia wash to prevent chemical reaction with uranium. The base of the mold cavity where solidification starts was given a thin zircunia wash, in keeping with normal uranium casting practice. This coating prevents chemical reaction between the graphite and the uranium. The thickness of the casting was determined by the requirement that solidification not be affected by radiative heat loss from the top of the casting. That is, heat extraction from the casting is primarily downwards into the mold. initial mold temperature, 600°C, was chosen to match normal uranium casting temperatures practice. Mold were measured with chromel-alumel thermocouples. Metal temperatures were measured with platinum - platinum + 10% rhodium thermocouples encased in tantalum tubes. One experiment also used exposed platinum thermocouples. Although the exposed thermocouples were chemically attacked by the uranium, they did indicate that the tantalum sheathing did not introduce serious errors in measuring Mold preheating was accomplished by induction solidification times. heating which gave a total temperature differential across the mold of less than 10°C. The experiments were performed in a vacuum induction furnace equipped with separate coils to heat crucible and mold.

No Superheat

The analytical approach of the Virtual Adjunct Method requires the superheat to be negligible. In spite of the practical difficulties of handling molten uranium, it was possible to introduce the metal into the mold at a superheat which varied from about 12° to 0°C. The variation in superheat is illustrated in Fig. 5 which shows that the peak temperature reached by the metal nearest the interface was  $11\overline{45}$ °C.



Figure 5. Temperature versus time plot for thermocouples placed 33 mm and 55 mm from the metal/mold interface, solid and dotted lines respectively. The data indicates some superheat in the metal near the interface but none in the metal further away.

The thermocouple remote from the interface indicated a maximum temperature equal to the melting point, 1132°C. The results of the experiment are plotted in Fig. 6 on the same type of plot as used for Fig. 1.



Figure 6. Freezing time over distance plotted versus distance from interface for uranium solidifying in a graphite mold. Squares are experimental points and asterisks are simulated points. Solid line is calculated from the VAM theory. Both theory and simulation were calculated with an interfacial heat transfer coefficient of

The best fit for an interface heat transfer coefficient was  $2 \text{ kW/m}^2/\text{K}$ . Again a simulation using ABAQUS followed the theory very closely except that the inclusion of radiative heat transfer across the gap elements appeared to have no effect on the effective interface coefficient. Temperature versus time histories are displayed for two locations, one in the mold 50 mm from the interface and one in the metal 43.5 mm from the interface, as shown in Figs. 7 and 8 respectively. Good agreement between experiment and simulations was obtained.



Figure 7. Temperature versus time plot for a thermocouple placed in the mold 50 mm from the interface. Solid ard dashed lines show experimental and simulated histories, respectively. Figure 8. Temperature versus time plot for a thermocouple placed in the metal 43.5 mm from the interface. Solid and dashed lines show experimental and simulated histories, respectively.

Radiative heat loss from the top of the casting leads to the formation of an appreciable thickness of solidified metal, independent of the solidification due to the mold. Simulation shows that the last metal to freeze is approximately 22 mm down from the top of the casting as illustrated in Fig. 4. In the actual casting this is revealed by large shrinkage cavities at this location.

### Superheat Present

An experiment was run with the metal at the normal superheat of 200°C, for comparison with normal casting practice. The pouring temperature was 1330°C. The type of plot used in Fig. 1 is not appropriate for this siutation. Instead, the freezing time is plotted versus the distance from the interface, Fig. 9.



Figure 9. Freezing time versus distance from interface plotted for superheated uranium solidifying in a graphite mold. Asterisks show experimental points. Lines are simulated data for interface heat transfer coefficients of 1, 2 and 5 kW/m<sup>2</sup>/K. Longer freezing times in the simulated data are associated with lower heat transfer coefficients.

The results show the characteristic features of a delay in the start of solidification followed by progress of the freeze front, first as it accelerates and then as it decelerates. A finite element simulation is useful for this situation where no explicit analytical form is available. Also plotted are three simulations where the interface heat transfer coefficient was assigned values of 1, 2 and 5 kW/m<sup>2</sup>/K. As with the previous experiment the best fit with the experimental data was obtained for an interface coefficient of 2 kW/m<sup>2</sup>/K. Figure 10 compares the temperature time history for a location 100 mm from the interface in the graphite mold. Good agreement was also obtained for a location in the metal 44 mm from the interface, Fig. 11.



Figure 10. Temperature versus time plotted for a thermocouple placed in the mold 100 mm from the interface. Solid and dashed lines show experimental and simulated data respectively.



Figure 11. Temperature versus time plotted for a thermocouple placed in the metal 43.5 mm from the interface. Solid and dashed ines show experimental and simulated data, respectively. Increased thermal conductivity in the liquid was accounted for by assuming a factor of ten for the difference between solid and liquid uranium. The measurements of Harrison and Weinberg [6] on liquid tin were the basis for this assumption. As a check on the assumed thermal conductivity of liquid uranium, two further simulations were run. An interface coefficient of 2 kW/m<sup>2</sup>/K was used and the conductivity of the liquid uranium. Figure 12 compares experimental data with simulations with three different factors for the thermal conductivity in the liquid.



Figure 12. Freezing time versus distance from interface plotted for superheated uranium solidifying in a graphite mold. Asterisks are experimental points. Lines show simulations for the same interfacial heat transfer coefficient, 2 kW/m<sup>2</sup>/K, but with different liquid metal thermal conductivities. Solid, dotted and dashed lines show data for increases by factors of 2.5, 5 and 10 times the solid thermal conductivity, respectively.

The best agreement with the experimental data is given by the simulation which used a factor of 5 for the liquid metal conductivity. This estimate of effective liquid uranium thermal conductivity requires experimental verification, however. This is an important prameter for accurate solidification modeling of complex uranium shapes.

# Conclusions

Ar. effective metal/mold interface heat transfer coefficient of  $2kW/m^2/K$  was calculated from data for unalloyed uranium solidifying in a graphite mold. This compares favorably with the value of 1.2 kW/m<sup>2</sup>/K measured for an aluminum bronze by Ho and Pehlke [7].

The simulations using the ABAQUS finite element code were, in general, in good agreement with the experimental data.

The emissivity of an exposed surface of a uranium casting was estimated at 0.2. For an emissivity of 0.2, radiation should contribute less than 4% of the effective heat transfer coefficient.

The effective thermal conductivity of liquid uranium was estimated to be five times that of solid uranium. This can be compared with the factor of ten measured for liquid tin, which was used as an initial estimate for modeling uranium.

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# Variables

k <sub>s</sub>	thermal conductivity of solid 👞 ɔl,	47.8 W/m/K
k <sub>m</sub>	thermal conductivity of mold,	50.0 W/m/K, at 1000°C,
		100.0 W/m/K, at 20°C
d <sub>s</sub>	density of solid metal,	19 Mgm/m <sup>3</sup>
d <sub>m</sub>	density of mold,	1.6 Mgm/m <sup>3</sup>
c <sub>s</sub>	specific heat of solid metal,	160 J/kg .
c <sub>m</sub>	specific heat of mold,	1600 J/kg
s <sub>o</sub>	"virtual adjunct" or effective preexist	ing thickness of metal
s	actual thickness of solidification metal	l
t	time at which the metal temperature temperature at a given position,	passes through the liquidus
a <sub>s</sub>	thermal diffusivity of metal,	$1.6 \times 10^{-5} \text{m}^2/\text{s}$
Φ	dimensionless parameter of equation 4,	0.64
Μ	dimensionless parameter of equation 5,	0.8589
h	interface heat transfer coefficient, W	/m <sup>2</sup> /K
T <sub>o</sub> me	elting temperature of metal,	1132°C
T <sub>f</sub> ir	nitial temperature of mold,	600°C
L, me erf (	etal latent heat of fusion, $(\phi) = \sqrt{\frac{2}{\pi}} \int_{0}^{\phi} \exp(-x^2) dx$	38720 J/kg

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