1 Introduction

Fuel reprocessing is an integral part of the IFR (Integral Fast Reactor) prototype being constructed at Argonne National Laboratory (Till, 1989). Two of the prime safety features of this advanced power reactor is the on-site fuel reprocessing which prevents diversion of nuclear material after the plant is started up and the metallic fuel which allows electrochemical reprocessing and minimizes stored energy. Thus, the reprocessing is an advanced safety feature of this plant.

The IFR fuel reprocessing must be such that all criticality accidents are made incredible. This paper discusses how one type of criticality accident is made incredible in the fuel pin casting furnace. The feedstock for the furnace consists of solid pieces of metallic fuel placed in a crucible. The temperature of the feedstock is then raised to the melting point to produce the desired mixture of the materials for the cast fuel. The failure of the crucible under the worst conditions for forming a critical geometry was analyzed and design requirements formulated which eliminate any possibility of a critical geometry. A complete failure of the fuel casting crucible is not of concern because the material would spread out and solidify in a non-critical shape.

Small leaks from the crucible are of more concern even though several improbable events must occur in order for molten fuel to leak out of a crucible and travel down to the support plate to solidify into a critical geometry (see figure 1). Such a shape can be formed from a leak which continually impinges on the same spot. The process can be envisioned as the flow or drip of a liquid metal onto a flat surface with the metal solidifying as it transfers heat to the surface. After reaching the surface, the liquid flows out radially until surface tension stops it or it solidifies. The purpose of this study was to determine the shape, both experimentally and analytically, that a deposit takes for different temperature regions in the furnace, different support materials that the fuel can solidify on and different leak rates and to compare the model results to experimental measurements. The results of the study were used to select allowable furnace material which will prevent critical geometries from forming.

An analytical model was developed to describe the shape of the deposit. The model shows the shape is dependent on the leak (or droplet) flow rate, the liquid metal temperature, its solidification temperature, the substrate temperature, and the properties of both materials. Experiments were performed using a tin (95 antimony (5 conductor (copper), and the other a good insulator (concrete board). Comparisons between experiment and the analytical model were made and conclusions drawn.

2 Analytical Model

The determination of the shape of the deposit consists of three steps: 1) calculating the solid liquid interface height at each radial location, 2) determining the amount of deposit that is still liquid, and 3) distributing the liquid on the top of the solidified...
metal and substrate.

The solidification of liquid metal at each radial location is approximated by one dimensional heat conduction in the direction normal to the substrate. A pure metal (solidus temperature equals liquidus temperature) is assumed. An analytical solution which describes the temperature history when two semi-infinite materials are placed in contact is used to represent the solidification front motion. One material is the substrate, the other the liquid metal. The liquid metal at the center of the point of contact \( (r = 0) \) begins to transfer heat at time zero. Any liquid which is not solidified at time \( t \) is assumed to flow out to the liquid limit radius \( R(t) \) such that the thickness of the liquid \( \delta \) is uniform value for all \( r < R(t) \). The value \( \delta \) is determined by a balance between surface tension and the gravity force at the radius \( R \). Experimental observations show that this is a good approximation. The spread of the liquid metal is assumed circumferentially uniform. As liquid metal first reaches an as yet uncovered radial position, the initial time for the solidification front motion at that radial position is determined. The amount of metal that is still liquid is determined by summing the total material deposited to time \( t \) and subtracting from that the mass of solidified material.

### 2.1 Equations of Change for Solidification Problem

The one dimensional heat conduction problem as shown in figure 2 consists of three regions, the substrate (region 0), the solidified material (region 1), and the liquid (region 2). The analytical solution assumes the substrate region is infinite in the negative x direction and the liquid metal is infinite in the positive x direction. These are reasonable approximations in spite of them both being finite because: 1) The molten region is continually being resupplied by new molten material. 2) The substrate is thick enough (1 cm) such that the heat does not reach the bottom in the time of deposit if it is an insulator and conductive enough to act as if it is infinite in the x direction because it spreads heat throughout the whole plate if it is a good conductor like copper.

The conduction equation is the same in all three regions and is given by

\[
\rho_i C_p \frac{\partial T_i}{\partial t} = k_i \frac{\partial^2 T_i}{\partial x^2}
\]

where \( \rho_i, C_p, k_i, \) and \( T_i \) are the density, specific heat, thermal conductivity, and temperature in region \( i \).

The initial conditions are

\[
\begin{align*}
T_2(0, x) &= T_h \\
T_0(0, x) &= T_c
\end{align*}
\]

The boundary conditions at infinity are
\[ T_2(t, \infty) = T_h \]
\[ T_0(t, -\infty) = T_c. \]  

(3)

The interface location between the liquid and solid metal is defined as \( X(t) \). Two conditions are present at this interface

\[ T_1 = T_2 = T_m \]  

(4)

where \( T_m \) is the melt-solidification temperature and

\[ k_1 \frac{\partial T_1}{\partial x} - k_2 \frac{\partial T_2}{\partial x} = L \rho \frac{\partial X}{\partial t} \]  

(5)

where \( L \) is the heat of fusion and \( X \) is the location of the solid liquid front.

The boundary condition at the interface between the metal and substrate can either be represented as perfect contact, or as a resistance to the heat transfer at \((t, 0)\), respectively by

\[ T_0(t, 0) = T_1(t, 0) \]  

or

\[ k_1 \frac{\partial T_1}{\partial x} = h(T_1 - T_0) \]  

(6)

where \( h \) represents the interface heat transfer coefficient.

The solution with the latter boundary condition approaches that of the former as \( h \) approaches infinity. In either case, the heat flux at \((t, 0)\) must also be equal as

\[ k_0 \frac{\partial T_0}{\partial x} = k_1 \frac{\partial T_1}{\partial x} \]  

(7)

2.2 Analytical Solution for the Front Motion Problem

Shamsunder (1988) points out that the latent heat of fusion for metals is often negligible. If the heat of fusion of the liquid metal is neglected and the contact resistance assumed zero, an analytical solution for a combined region 1 and 2 is given in Carslaw (1959) as

\[ \Phi = \frac{\beta_1}{\beta_1 + \beta_0} \left\{ 1 + \frac{\beta_2}{\beta_1} \text{erf} \left( \frac{x}{2\sqrt{\alpha_1 t}} \right) \right\} \]  

(8)

where

\[ \Phi = \frac{T - T_c}{T_h - T_c} \]

\[ \beta_i = \sqrt{k_i \rho_i C_{pi}} \]
The progress of the solidification front can be tracked by setting the temperature to the melt temperature and solving for \( X(t) \). Rearranging equation 5 yields

\[
X = 2\lambda \sqrt{\alpha_1 t} \tag{9}
\]

where

\[
\lambda = \text{erf}^{-1} \left\{ \frac{\beta_1}{\beta_0} \left( \frac{\Phi_m}{\beta_1} \frac{\beta_1 + \beta_0}{\beta_1} - 1 \right) \right\}
\]

\[
\Phi_m = \frac{T_m - T_c}{T_h - T_c}
\]

To take into account the effects of the heat of fusion, this solution can be modified by raising \( T_h \) by a temperature equivalent. In this case, \( \delta \) would be replaced by

\[
\Phi_m = \frac{T_m - T_c}{T_h + \frac{c_p}{\Delta s} - T_c} \tag{10}
\]

An alternate solution, again from Carslaw (1959), which takes into account heat transfer contact resistance between the deposit and substrate as specified in boundary condition 6b is given as

\[
\Phi = \frac{\beta_1}{\beta_1 + \beta_0} \left\{ 1 + \frac{\beta_c}{\beta_1} \left[ \text{erf} \left( \frac{x}{2\sqrt{\alpha_1 t}} \right) + e^{h_1 x + h_2^2 \alpha_1 t} \text{erfc} \left( \frac{x}{2\sqrt{\alpha_1 t}} + h_1 \sqrt{\alpha_1 t} \right) \right] \right\} \tag{11}
\]

where

\[
h_1 = \frac{h(\beta_1 + \beta_0)}{k_1 \beta_0}
\]

The motion of the melt temperature cannot be solved for explicitly from this solution but can be obtained either by iteration or solving for \( \frac{dX}{dt} \) by combining the solution of equation 11 with the definition of the total derivative of temperature set to zero as

\[
dT = \frac{\partial T}{\partial x} dX + \frac{\partial T}{\partial t} dt \tag{12}
\]

Thus, from either equations 9 or 12, the function for the motion of the solidification front is given as \( X(t) \) assuming \( t \) is measured from the time the metal first contacts the substrate.

### 2.3 Shape of Solid Deposit as a Function of Time

The time at which liquid metal first reaches a radial location \( r \) is designated as \( t_f(r) \). This time is solved for from the conservation of mass equation where the total mass deposited until time \( t \), \( M_{in} \), is equated to
\[
\frac{M_{in}}{\rho_1} = 2\pi \int_0^{R(t)} x_s(r,t)r \ dr + \pi \delta R(t)^2
\]  

(13)

R(t) is solved for from this equation and inverted. The first term on the right hand side of this equation represents the volume in the solid, the second, the mass in the liquid. In order to arrive at the second term, the assumption is made that the vertical, \( x \), dimension of the liquid film is a constant, \( \delta \).

The solidification front \( x_s(r,t) \) is given by the \( X(t) \) solution as

\[
x_s(r,t) = X(t - t_f(r))
\]  

(14)

In general, the function \( t_f(r) \) is obtained as the inverse of the function \( R(t) \) determined in equation 13 as long as \( R(t) \) is monotonically increasing. For small leak rates, this function may have periods where it regresses. During periods when there is no liquid metal on top of the solid material at location \( r \), \( t_f(r) \) for all \( r > R(t) \) is increased by the amount of time it is uncovered.

### 2.4 Determination of Liquid Metal Film Thickness

Although the film thickness is a function of time and radius, the approximation is made that the vertical dimension of the film thickness is constant and determined by a force balance between gravity and surface tension at \( R(t) \). Gravity tends to make the film uniform on near horizontal surfaces and the amount of fluid contained in the film very small on steep surfaces. Both these effects are consistent with the assumption of a constant vertical dimension. The surface tension force is small except near the edge \( R(t) \). The force due to surface tension is approximated by that of the calculated pressure difference between the inside and outside of a long cylinder of liquid. One half of such a cylinder approximates the shape of the outside edge of the liquid film. As in Sears (1955), a force balance yields the pressure difference between the inside and outside of the film as

\[
2 L \sigma = (P_i - P_o)2 \ R \ L
\]  

(15)

where

- \( L \) = the length of the cylinder.
- \( \sigma \) = the surface tension.
- \( R \) = the radius of curvature.
- \( P \) = the pressures on the inside and outside.

The pressure in the film is approximated by the gravity force on the substrate surface.
\[ P_i = P_a + \rho g h \]  \hspace{1cm} (16)

Equating the radius in Equation 14 with half the film height and substituting equation 15 for the pressure difference gives

\[ \delta = \sqrt{\frac{2\sigma}{\rho g}} \]  \hspace{1cm} (17)

This value is used for the uniform film vertical height used in Equation 13. Hocking (1992) presents a much more detailed modeling of a single droplet which takes into account the force due to wetting.

### 2.5 Model Limitations

Besides the approximations mentioned above, the major limiting assumptions of the model are 1) the neglect of radial conduction and 2) the neglect of heat transfer from the upper surface of the molten metal to the surroundings. The first limitation causes the model to overpredict the center height which is acceptable since we desire designs which produce flat deposits. The second limitation, as will be seen later, causes low values of center height to be predicted for deposits on insulators. This is because the heat transferred to the surroundings is of the same order of magnitude as to the substrate when the latter is an insulator and small when it is a conductor.

### 3 Analytical Conclusions and Predictions

The analytical model predicts that the deposit shape is dependent on the groups, \( \dot{V}, \Phi, \beta, \alpha_1 \), and \( h \). \( \dot{V} \) is the volumetric leak rate related to \( M_{in} \), the liquid density, and the time. In all cases studied, \( \dot{V} \) was taken as a constant so that

\[ M_{in} = \dot{V} \rho t \]  \hspace{1cm} (18)

It is important to note that for constant properties, the center height, \( H = X(t) + \delta \), (determined from equation 9 or 12), is predicted to be independent of \( \dot{V} \) and dependent only on the total time of the deposit. This implies that for a given total volume of deposit, the shape will candle for low flow rates and pancake for large flow rates. Somewhere in-between these two shapes is the unwanted most compact or hemispherical shape. The center height increases with increasing heat transfer to the substrate, that is increasing \( \beta, \alpha_1 \), or \( h \). It also increases the closer the liquid metal temperature is to the melting temperature, that is, increasing \( \Phi \).

The minimum leak rate of concern is bounded by the amount of metal fuel available and the crucible heat up rate. The maximum plutonium allowed in the crucible is 4 kg. Plutonium melts at 900\( K \) and uranium at 1400\( K \). When both materials are molten,
they mix and the neutron multiplication factor decreases so that criticality is no longer of concern. Neglecting the dissolution of solid uranium in liquid plutonium, the heat up time from 900 to 1400$K$ is taken as the maximum time for the leak. This yields a leak volume of 4 kg/18 g/cc = 222.2 cc and a minimum leak rate of 0.37 cc/sec. A leak rate lower than this would produce a deposit of plutonium diluted by uranium and not a concern for criticality.

3.1 Deposition on a Conductor

Figure 3 shows the calculated shape result for various times for a leak rate of 0.3 cc/sec for deposition of plutonium on copper with no interface resistance. The entire 4 kg of plutonium is deposited in 740.7 sec and the final shape is seen to have a 7/3.6 $H/R_b$ ratio where $R_b$ is the boundary radius of the deposit. The early shapes (before 60 sec) have $H/R_b$ ratios less than 1.

Figure 4 contains results for a high leak rate and is seen to pancake for all times (final $H/R_b = 1.3/8.3$). Figure 5 shows that an intermediate leak rate of 1 cc/sec produces a final shape which is near hemispherical (final $H/R_b = 3.9/4.8$) even though the early shapes are flatter.

The effect of the liquid film thickness is shown in Figure 6 where $\delta$ is taken to be 0.2 cm whereas all the other results in this section used 0.1 cm. The results are for the highest leak rate, $V=10$ cc/sec, and should be compared to figure 4. The effect is seen to be small (final $H/R_b = 1.4/7.9$ instead of 1.3/8.3) and will be smaller for lower flow rates. The calculated value of $\delta$ for $Pu$ from equation 17 is approximately 0.22 cm.

Liu (1992) indicates that the interface heat transfer coefficient should be between 0.1 and 1.0 W/cm$^2$ K for single drops dropped 26.5 cm whereas Watanabe assumed an infinite heat transfer coefficient. The effect of the interface resistance is shown in figures 7 and 8 for $h = 1$ and $h = 0.1$ W/cm$^2$ K respectively. The results for $h = 1$ are almost identical to that of $h = \infty$, figure 5. The results for $h = 0.1$ shows a twenty percent decrease in the center height and a ten percent increase in the radius. Thus the conclusion is reached that in the range of expected parametric variation, the conclusion about criticality would not change much. The results for $h = 0.1$ does exhibit a behavior not shown in the other results. The other results exhibited a monotonic increase in the liquid radius with time. For the $h = 0.1$ case, the liquid radius reaches a local maximum near 6 sec., retreats until 12 seconds, then increase again beyond that time. The shape at 20 seconds specifically shows the shape resulting from this behavior.

The above results show that copper is not an acceptable substance to use as a substrate in the casting furnace.

3.2 Deposition on an Insulator

An interesting behavior of the analytical model for $h = \infty$ is that the argument in equation 9 must be positive or the material will not solidify. This behavior is observed
because ambient heat loss has been neglected. Thus, for solidification,

\[ \Phi_m = \frac{T_m - T_c}{T_h - T_c} > \frac{\beta_1}{\beta_1 + \beta_0} \]  \hspace{1cm} (19)

must be satisfied. In some cases, this may represent real behavior early in the transient such as when a liquid metal on a metal of the same material and implies that the liquid metal temperature is high enough that the substrate will melt rather than the liquid solidify. The solution given in equation 9 also implies for \( h = \infty \) that the interface temperature remains constant for all time.

Due to the above limitation, in order to obtain a solution for Pu depositing on an insulator, the liquid temperature had to be taken close to the solidification temperature. Figure 9 shows the deposit shape for the minimum leak rate. The plutonium "pancaked." Higher leak rates would produce flatter shapes. This leads to the conclusion that if an insulator instead of a conductor is used in all places where a deposit is possible the material will spread out rather than produce a critical shape.

4 Comparison to Experimental Data

A series of experiments were run with a common solder material (955 of using plutonium and high temperature.

4.1 Experimental Apparatus and Procedure

The experimental apparatus is shown schematically in figure 10. It consists of a solder melting pot with a valve in the bottom. The temperature of the molten metal was controlled with a thermocouple driven power unit. The substrate was positioned with a lab jack. After temperature equilibrium was attained, the valve was opened to allow an estimated drip rate by hand. The valve was adjusted manually throughout the test to maintain a uniform drip rate since the liquid metal driving head was decreasing. Flow rates were computed by dividing the total mass deposited by the density and duration of the test. Flow rates may have varied as much as 20 percent during an individual test. The lab jack was adjusted to maintain an approximate 4 cm. drop height to prevent splashing. Splashing tends to lower the maximum deposit height so would produce results inconsistent with the model. Dimensional measurements were made with a hand held ruler. Height is judged accurate to $\pm 0.2$ cm., width to $\pm 0.5$ cm.

4.2 Experimental Results

The height measurements for all experiments are shown in figure 11 and are seen to separate into three groups: 1) the near melting point group on copper, 2) the high temperature group on copper, and 3) a near melting point group on an insulator.
Analytical height predictions are supplied for the first two groups using zero contact resistance and the modified $T_m$ group given in equation 10, and are seen to agree well. The model predicts no solidification for the metal on the insulator so the height is zero for all time. As mentioned previously, this latter difference is due to neglect of heat transfer from the upper surface and will be the subject of further investigation. The build up is small in the case which was run at a leak rate less than half that of the minimum (0.17 cc/sec) so the height would be large enough to measure. A leak rate of 0.37 cc/sec would produce an even flatter shape.

Figure 12 concentrates on the results from the near melting point tests on copper to show leak rate as a parameter. The liquid metal temperature, $T_h = 260K$, was the same for all the tests. The experimental results demonstrate the analytical conclusion that the center height as a function of time is independent of leak rate. One test was run to test the effect of drop height (15 cm.) and it produced the highest center heights (see t19). The higher center height of this test may be due to additional loss of heat during the drop causing the liquid metal to be at a lower temperature at contact. No splashing occurred as had been observed in some earlier preliminary tests and may be a surface tension effect or because the temperature was so close to solidification.

Figure 13 shows the results for the high temperature liquid metal depositing ($T_h = 460K$) on copper. As predicted by analysis, the higher temperature case shows a much lower build-up of material. Five tests and a prediction are shown in this figure. A single endpoint measurement were made for three of the tests. Single point tests t1 and t2 fall on the same curve as multipoint test t18 and above the analytical prediction. Test t17 falls below the prediction. Single point test t0 was at a lower metal temperature ($T_h = 427K$) than the other tests and prediction so should produce a higher height. Again, the theory between experiment and theory is good.

5 Conclusions

An analytical model has been developed to predict the shape of a deposit from a liquid metal leaking (dripping) onto a substrate and solidifying. Experimental results were obtained which showed good agreement with the model. The model predicts and the data confirm that a problem hemispherical shape, from the standpoint of criticality, can develop if the deposit is onto a metal substrate. Conversely, the shape will be short and squatty if the material deposits on a good insulator and therefore not a criticality concern. Future work will involve including ambient heat loss so that the shape depositing on an insulator can be accurately predicted for the specific conditions in a nuclear fuel casting furnace.

DISCLAIMER

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References


Table 1: Material Properties Used in Calculations.

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<th></th>
<th>Pu</th>
<th>Sn'</th>
<th>Cu</th>
<th>Composite²</th>
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<tbody>
<tr>
<td>$T_m$ (°C)</td>
<td>640</td>
<td>240</td>
<td></td>
<td></td>
</tr>
<tr>
<td>L (J/gm)</td>
<td>11.8</td>
<td>59.5</td>
<td></td>
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<tr>
<td>k (W/cm K)</td>
<td>0.1</td>
<td>0.6</td>
<td>3.8</td>
<td>0.0104</td>
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<tr>
<td>$\rho$ (gm/cc)</td>
<td>18.0</td>
<td>7.1</td>
<td>8.95</td>
<td>0.8</td>
</tr>
<tr>
<td>$C_p$ (J/gm K)</td>
<td>0.167</td>
<td>0.244</td>
<td>0.42</td>
<td>1.68</td>
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Table 2: Temperatures Used in Calculation.

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<th>$T_m$ °C</th>
<th>$T_c$ °C</th>
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</thead>
<tbody>
<tr>
<td>Plutonium on Cu</td>
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<td>640</td>
<td>133</td>
</tr>
<tr>
<td>Plutonium on Composite</td>
<td>657</td>
<td>640</td>
<td>133</td>
</tr>
<tr>
<td>Solder on Cu (low T)</td>
<td>260</td>
<td>240</td>
<td>24</td>
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<tr>
<td>Solder on Cu (high T)</td>
<td>460</td>
<td>240</td>
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Note:
1 - Properties of 95 Tin 5 Antimony used for $T_m$, rest were for Tin.
2 - Wood composite softboard properties estimated.
Figure 1: Casting Furnace Schematic.
Figure 2: One-Dimensional Solidification Model.
Figure 3: Shape of Deposit on Conductor 0.3 cc/sec.
Figure 4: Shape of Deposit on Conductor 10. cc/sec.
Figure 5: Shape of Deposit on Conductor 1.0 cc/sec.
Figure 6: Deposit on Conductor 10. cc/sec $\delta = 0.2$ cm.
Figure 7: Deposit on Conductor 1.0 cc/sec h=1 w/cm2K.
Figure 8: Deposit on Conductor 1.0 cc/sec h=.1 w/cm2K.
Figure 9: Deposit on Insulator 0.3 cc/sec.
Figure 10: Schematic of Equipment.
Figure 11: Experimental Height Measurements.
Figure 12: Near Melt Temperature on Conductor.
Material = 95 Sn 5 Sb

T melt = 860 F
Copper Chill Plate

Figure 13: High Liquid Temperature on Conductor.
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