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USING CENTER HOLE HEAT TRANSFER TO REDUCE FORMATION TIMES FOR CERAMIC WASTE FORMS FROM PYROPROCESSING

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ABSTRACT

The waste produced from processing spent fuel from the EBR II reactor must be processed into a waste form suitable for long term storage in Yucca Mountain. The method chosen produces zeolite granules mixed with glass frit, which must then be converted into a solid. This is accomplished by loading it into a can and heating to 900 C in a furnace regulated at 915 C. During heatup to 900 C, the zeolite and glass frit react and consolidate to produce a sodalite monolith. The resultant ceramic waste form (CWF) is then cooled. The waste form is 52 cm in diameter and initially 300 cm long but consolidates to 150 cm in length during the heating process. After cooling it is then inserted in a 5-DHLW/DOE SNF Long Canister. Without intervention, the waste takes 82 hours to heat up to 900 C in a furnace designed to geometrically fit the cylindrical waste form.

This paper investigates the reduction in heating times possible with four different methods of additional heating through a center hole. The hole size is kept small to maximize the amount of CWF that is processed in a single run. A hole radius of 1.82 cm was selected which removes only 1% of the CWF. A reference computation was done with a specified inner hole surface temperature of 915 C to provide a benchmark for the amount of improvement which can be made. It showed that the heatup time can potentially be reduced to 43 hours with center hole heating.

The first method, simply pouring high temperature liquid aluminum into the hole, did not produce any noticeable effect on reducing heat up times. The second method, flowing liquid aluminum through the hole, works well as long as the velocity is high enough (2.5 cm/sec) to prevent solidification of the aluminum during the initial front movement of the aluminum into the center hole. The velocity can be reduced to 1 cm/sec after the initial front has traversed the ceramic. This procedure reduces the formation time to near that of the reference case. The third method, flowing a gas through the center hole, also works well as long as the product of heat capacity and velocity of the gas is equivalent to that of the flowing aluminum, and the velocity is high enough to produce an intermediate size heat transfer coefficient. The fourth method, using an electric heater, works well and heater sizes between 500 to 1000 Watts are adequate. These later three methods all can reduce the heatup time to 44 hours allowing production to be doubled and a more uniform heating.

INTRODUCTION

Argonne National Laboratory has developed a high temperature electrochemical method for extracting plutonium and minor actinides from spent nuclear fuel which can then be used as feedstock for fast breeder reactor fuel.[1] The fission products remaining in the electrolyte must then be stored as nuclear waste. The pyroprocessing removes the long lived transuranics (100,000 year storage if treated as waste) and leaves behind fission products (including I-129 and Tc-99) in the elecrtrorefiner electrolyte (only 300 year storage needed). The process analyzed here is used to convert the fission products into a safe form for Yucca Mountain storage. This form reduces the volume to one fifth that of the original spent fuel. Removal of the transuranics reduces the decay heat by one fifth as well so the capacity of the Yucca Mountain repository is effectively expanded by a factor of five. The much shorter storage time requirement would also make licensing of the repository much easier.

The spent fuel treated (metal fuel) is from the Experimental Breeder Reactor (EBR-II), but the process is being expanded to Light Water Reactor spent fuel (oxide fuel). The electrolyte waste stream[2] consists of a LiCl-KCl eutectic containing alkali, alkali earth, and lanthanide fission products present as chloride salts. To make the ceramic waste form, these halides are first occluded into the pores of zeolite-4A using an elevated temperature blending process. This salt loaded zeolite[3] is then mixed with glass frit (which is

crushed/ground/powdered glass). and loaded into cylindrical stainless steel canisters. Finally, the zeolite is consolidated by heating to 900 C and then cooled. The resulting ceramic waste form (CWF) is a glass-bonded sodalite[4] suitable for long-term storage in a repository. The heatup step is analyzed here.

The waste form size is 52 cm in diameter and initially 300 cm long but consolidates to 150 cm to be ultimately stored in a DOE canister (labeled the 5-DHLW/DOE SNF Long Canister). Experiments on this waste form are currently being conducted at the Material Fuel Complex (MFC) facility at the Idaho National Laboratory (INL). Shorter forms have already been successfully produced. A full length cylinder will be constructed in the near future.

The purpose of this paper is to investigate the potential benefits of center hole heat transfer in reducing the heat up formation times for the CWF and decreasing product non-uniformity. The material can consolidate unevenly and form a dome-shaped top. A heavy plate on the top of the waste form keeps it in a cylindrical shape and must be dealt with in providing a center hole for assisting in heat transfer and would discourage the use of multiple holes. The CWF is heated in a 915 C furnace to 900 C to densify (factor of 2) and bond the waste into a stable sodalite monolith. The heatup must be somewhat uniform to prevent non-uniform CWF densification. The methods used to assist in the heatup should not raise the temperature of any CWF portion above 970 C.

Approximately 40 runs will be needed as a result of processing EBR-II fuel, but the time reduction still significantly reduces costs in future years. It also serves as a model for a 100 MT/yr reprocessing plant for processing spent fuel from commercial nuclear plants. Center hole heating should heat the CWF more uniformly resulting in a more uniform densification and a more accurate density measurement.

Several methods of heatup are investigated including filling the hole with liquid aluminum, flowing liquid aluminum or gas through the hole, and inserting a rod heater in the hole. A previous reference[5] investigated the benefit of using solid metal rods to enhance the heat transfer and showed the surprising result that a steel rod slowed down the heating.

Each of these methods has potential advantages and disadvantages. The first, filling the center tube with liquid aluminum is advantageous because it involves pouring a heated liquid which will use its heat capacity to heat to the ceramic and has the potential to assist in the cool down phase[5]. The second method, flowing liquid aluminum, has the advantage of supplying a constant high temperature heat source through the center hole. Aluminum and lead are possible candidates because they are both liquid in the range of interest. Aluminum was selected because of its high thermal conductivity (237 W/m K), low melting temperature (660 C), high boiling temperature (2450 C), and large thermal capacity (density times specific heat, 2.44 J/cm^3 K). Lead has a much lower thermal conductivity and somewhat lower thermal capacity. (Reference 5 which lists most of the properties used here.). The third, flowing high temperature argon through the center hole, has the advantage of being able to easily switch to low temperature air to assist the cooldown. The fourth, using a heater rod, would supply heat during the heatup phase but then would not be useful for the cooldown phase.

Reference Calculation: In order to evaluate these methods, a reference calculation was made using a finite difference solution to the cylindrical heat conduction equation. The temperature profiles in the ceramic waste form with a hole in the center with the 915 C temperature impressed on the inside of the hole, as well as all other surfaces, serves as this reference. The temperatures calculated on the midplane at different radii are shown in Figure 1. Since this is a symmetric problem, the minimum temperatures are on the midplane. The minimum ceramic temperature, r_{min}, also included, reaches 900 C in about 43 hours so that the entire CWF form has reached or exceeded the target temperature at this time.

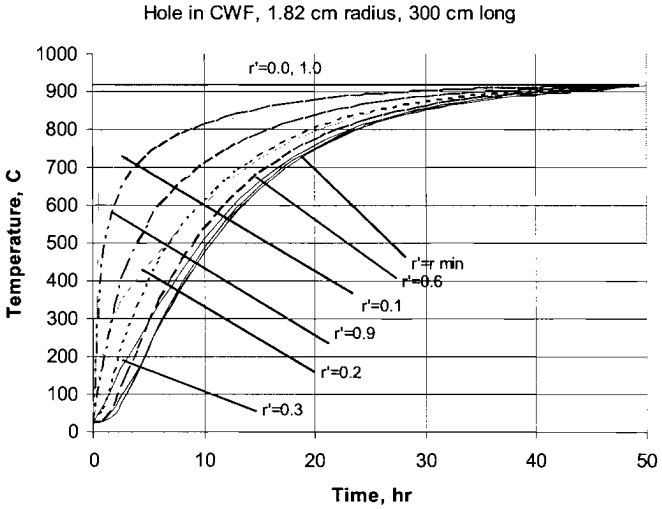


FIGURE 1. REFERENCE CALCULATION – ALL TEMPERATURES REACH 915 C IN 43 HOURS.

ANALYTICAL MODEL

The CWF density, thermal conductivity, and specific heat increase during heatup and consolidation. Heatup was modeled with the heat conduction equation. For efficacy, constant properties at the initial density, thermal conductivity and specific heat were used. Use of constant properties allows the solutions to be applied readily to changed conditions.

The boundary conditions for the temperature (T) for the heatup are T_{init} for the initial temperature (25 C), and T_{surr} for the surrounding temperature for all surfaces (915 C furnace). The heat transfer coefficient during heatup is assumed infinite, so T_{surr} is the surface temperature. The independent variables include r for the radial coordinate, z for the axial coordinate, and t for time. The dimensionless temperature T' is defined as

$$T' = \frac{T - T_{surr}}{T_{init} - T_{surr}}$$

The conduction equation can then be written as:

$$\frac{1}{\alpha}\frac{\partial T'}{\partial t} = \frac{1}{R^2}\frac{1}{r'}\left(\frac{\partial}{\partial r'} r' \frac{\partial T'}{\partial r'}\right) + \frac{1}{L^2}\frac{\partial^2 T'}{\partial z'^2}$$

$$where\ \alpha = \frac{k}{\rho C_p} \quad r' = \frac{r}{R} \quad and \quad z' = \frac{z}{L}$$

k = thermal conductivity, ρ = density, and C_P = specific heat.

Multiplying through by R^2 yields

$$\frac{\partial T'}{\partial t'} = \frac{1}{r'}\left(\frac{\partial}{\partial r'} r' \frac{\partial T'}{\partial r'}\right) + \frac{R^2}{L^2}\frac{\partial^2 T'}{\partial z'^2}$$

$$\text{where } t' = \frac{t\alpha}{R^2} \text{ and } \frac{R}{L} \text{ are independent parameters}$$

The center tube or hole in the waste form can be used for various methods of heating. For flowing liquid aluminum through the center hole, the conductivity of the aluminum is so much greater than that of the ceramic waste form that the radial temperature profile in the aluminum is flat across the tube. The energy balance of the center tube with flowing aluminum at velocity V assuming constant properties is given as

$$\frac{\partial T_m}{\partial t} + V\frac{\partial T_m}{\partial z} = \alpha_m \frac{\partial^2 T_m}{\partial z^2} + \frac{2}{R_i}\alpha_{cm}\frac{\partial T_c}{\partial r}_{r=r_i}$$

$$\text{where } m = \text{metal} \quad c = CWF \quad r_i = \text{hole radius}$$

$$\alpha_m = \frac{k_m}{\rho_m C_{Pm}} \qquad \alpha_{cm} = \frac{k_m}{\rho_c C_{Pc}}$$

The boundary condition between the metal flow and the CWF for an infinite heat transfer coefficient is

$$T_m = T_c \quad at\ r = r_i$$

The boundary condition for a finite heat transfer coefficient is

$$-h_{mc}(T_m - T_c) = k_c \frac{\partial T_c}{\partial r}_{r=r_i}$$

where h_{mc} is the heat transfer coefficient between metal and CWF

If instead of a flowing fluid, the heating is done with a heater rod of the same radius as the tube, the boundary condition on the ceramic would be replaced by

$$q = k\frac{\partial T_c}{\partial r}_{r=r_i} \quad \text{where the specific heat flux } q \text{ is given by}$$

$$q = \frac{Q}{2\pi r_i} \quad \text{where } Q \text{ is the total heat rate to the heater rod.}$$

HEATUP OF THE CERAMIC WASTE FORM

In reference 5, the improvement in heat transfer which results from inserting an aluminum rod in the hole was evaluated. The temperature in that case reaches 900 C in 65 hours as shown in Figure 2. This compares to 43 hours minimum in the reference calculation (Figure 1) by keeping the walls of the hole at 915 C. The all ceramic (i.e., without a center hole) reaches 900 C in 82 hour. Thus, the improvement by inserting an aluminum rod in the ceramic is 17 hours (82-65 = 17). However, by keeping the inner wall temperature at 915 C, it is theoretically possible to reduce the time by another 22 hours (65-43 = 22), for a total possible improvement of 39 hours (82-43 = 39), a reduction of almost half.

The aluminum rod results shown in Figure 2 show the rod (r'=0) to climb just a small amount ahead of the minimum temperature ($r=r_{min}$). Thus, the aluminum rod helps, but it does not heat the inner ceramic surface up as quickly as Figure 1 shows is theoretically possible.

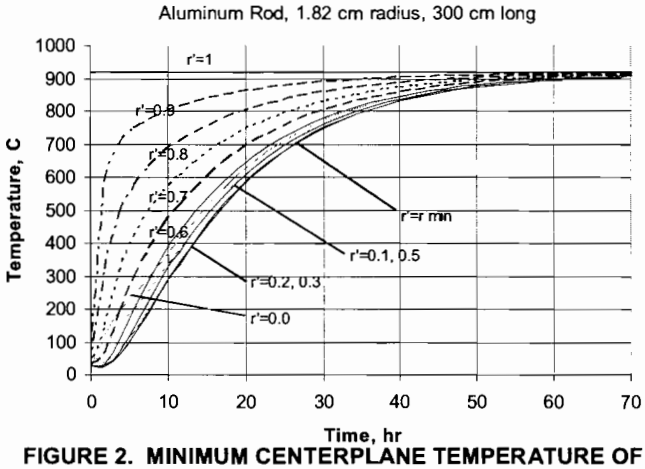


FIGURE 2. MINIMUM CENTERPLANE TEMPERATURE OF THE CWF WITH A SOLID ROD.

Pouring liquid aluminum into the center hole

The first method investigated was to pour liquid aluminum into the center hole. This was conceptualized by assuming the center hole was instantly full of liquid aluminum at a specified temperature at time zero. Thus, it was modeled with the above equations, setting V = 0 and using an infinite heat transfer coefficient between the metal and the CWF.

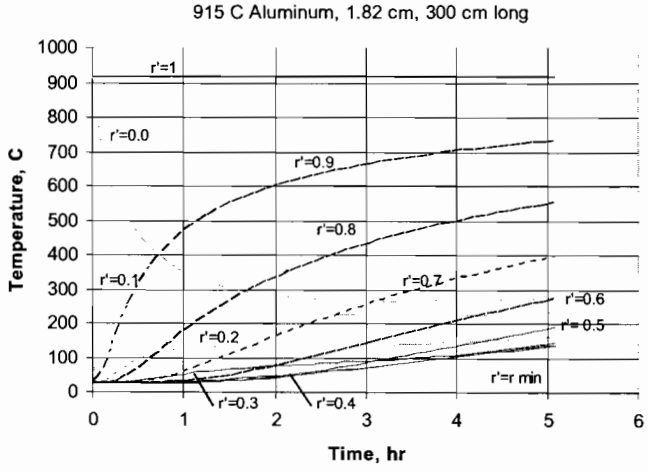


FIGURE 3 EFFECT OF 915 C ALUMINUM IN THE CENTER HOLE.

Short term results are shown in Figure 3 for aluminum being poured into the hole at an initial temperature of 915 C. It is seen that the temperature of the aluminum (r'=0) quickly diminishes to close to that of the CWF just inside the surface at r'=0.1. (Note, the interface between the aluminum and the ceramic is at r'=0.07). Thus, the aluminum has little effect because the rod is so small compared to that of the ceramic. The thermal capacity of the aluminum is larger at 2.44 J/cm^3 K

compared to that of the CWF which, is 0.74 J/cm^3 K but the volume of the CWF is so much larger.

The fact that the heat capacity in the aluminum is so small makes the long term results with liquid aluminum (initial temperature, 915 C) close to the aluminum rod results (Figure 2, initial rod temperature, 25 C) with small initial differences near the aluminum. The minimum temperature of the ceramic reaches 915 C virtually at the same time as that in Figure 2.

Since aluminum doesn't boil until 2450 C, another calculation was done with an initial aluminum temperature of 2000 C. The radial temperature profiles at the midplane are shown in Figure 4. The radial profiles show a clear effect of the initial temperature of the aluminum. (Note, the profile at t=0 is cut off to better show the rest of the temperature profiles but did start at 2000 C.) Within 10 hours, the aluminum has solidified and decreased to 400 C. The ends of the rod continue to conduct heat in from the furnace so the rod temperature continues to stay above the CWF temperature; however, as with the 915 C case, the long term results were almost identical to the aluminum rod without preheating (Figure 2). The minimum CWF reached 900 C in about 64 hours, just slightly less than a room temperature rod (65 hours), so that it is concluded that this method offers no benefit over the insertion of a room temperature solid aluminum rod.

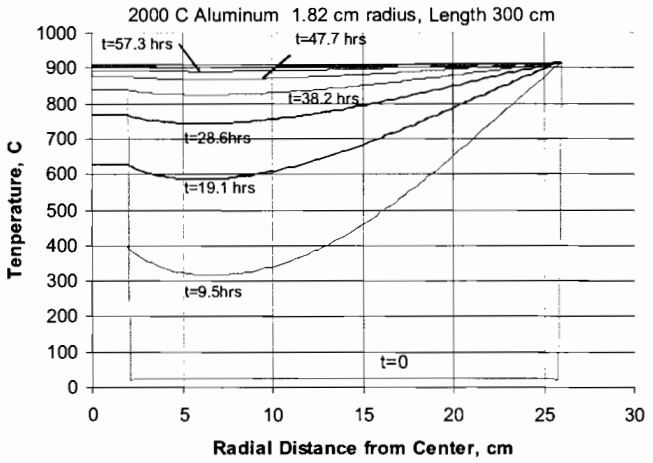


Figure 4 Radial Midplane Temperature Profile for Insertion of 2000 C Aluminum.

Flowing liquid aluminum through the center hole

The second method investigated to bring the ceramic up to temperature was to flow 915 C liquid aluminum through the center hole from the bottom. The hole is initially filled with argon gas. Rather than using a flow loop, the aluminum would be heated in a cylinder fitted with a piston that would push the aluminum up through the center hole into a cylinder on the top capable of heating the liquid back to 915 C. When the lower vessel is expended, the flow will be reversed and the process repeated as many times as necessary.

The solution for aluminum flowing at 1 cm/sec, with an infinite heat transfer coefficient between aluminum and CWF and an inlet temperature of 915 C, is shown in Figure 5. A velocity of 1 cm/sec will fill the 300 cm long hole in 300 seconds or 5 minutes. The calculation shows that the minimum temperature surpasses 900 C at approximately 43 hours or about the same as the reference case. Although the minimum temperature occurs higher up in the CWF than the midplane, the plot of the minimum temperature is also included in this figure.

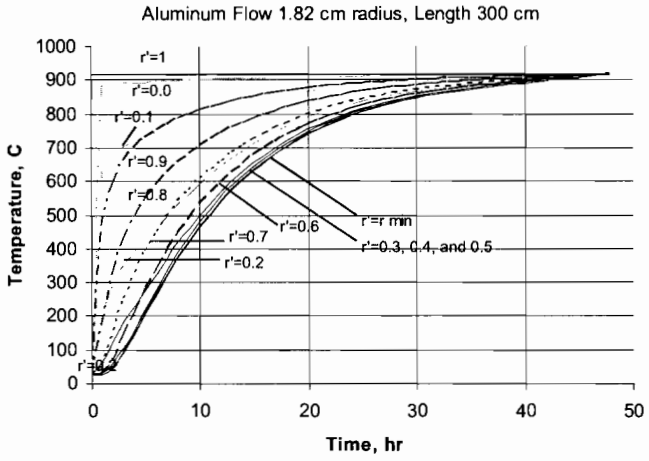


FIGURE 5 CWF HEATUP 915 C ALUMINUM FLOWING AT 1 cm/sec.

At 40 hours, the temperature is 889 C. The reference case is only 0.2 C higher at that time. Thus, this method seems to satisfy the requirements set up at the beginning. The radial temperature profile at the midplane is shown in Figure 6. The aluminum in the center hole appears to be at 915 C for the majority of the transient which is responsible for the good heatup performance. However, this temperature is not high enough in the early transient.

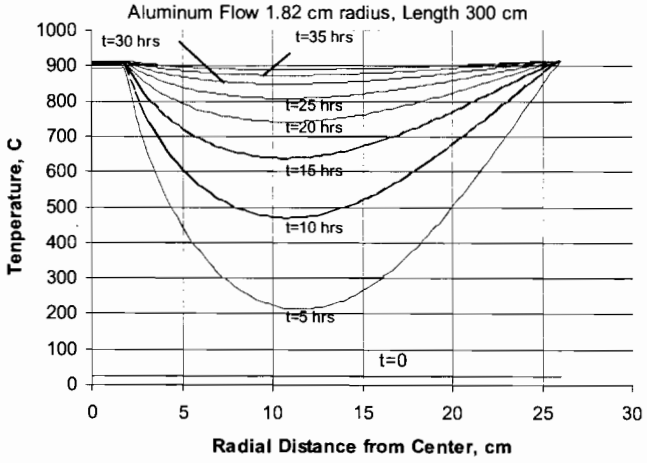
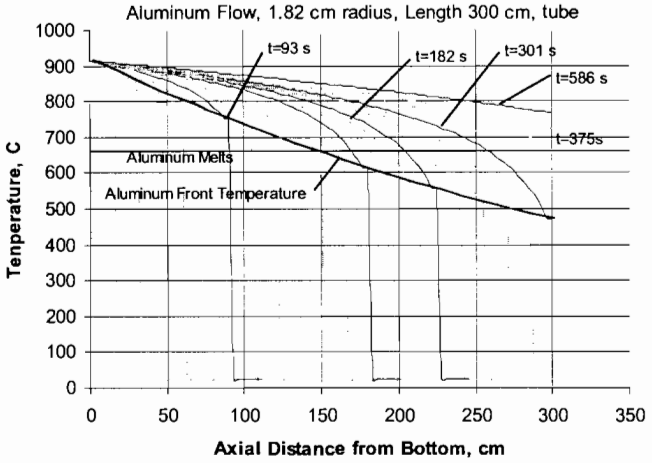


FIGURE 6 MIDPLANE RADIAL TEMPERATURE PROFILES 915 C ALUMINUM FLOWING AT 1 cm/sec.

The initial traverse of the liquid aluminum exposes the front of the flowing aluminum to a temperature of 25 C at each location in the tube. This extracts so much heat that the initial front of the aluminum solidifies and plugs the hole. The temperatures calculated for the initial front moving up the tube neglecting metal solidification is shown in Figure 7. It is seen that the front dips below the solidification temperature about halfway up the tube. Thus, the solution is no longer valid beyond this point. However, it is seen that if the aluminum could go beyond this point somewhat after the front exits the tube (~375 sec.), the heat loss decreases enough that the aluminum can pass through the tube without solidifying. The heat transfer coefficient used in this calculation (h = $10^5 W/m^2$ K) is large enough that the interface temperature calculated between the aluminum stream and the CWF is the same as the aluminum temperature.



FGURE 7 ALUMINUM FRONT TEMPERATURE DROPS BELOW SOLIDIFICATION.

A comment about the numerical solution: In order to keep the front from unrealistically spreading out, the conduction of heat in the axial direction in the metal was set equal to zero to calculate the front temperature. This underpredicts the front temperature because argon gas is transferring heat from the front and the liquid aluminum behind the front conducts heat to the front. The latter is larger than the former because of the ratio of conductivities (237 W/m K compared to 0.0162 W/m K). After the initial fill, this term is small. Runs with and without this term produced the same long term solution.

There are two methods which can be used to alleviate the problem. The velocity can be increased or a heat transfer resistance can be added between aluminum and ceramic.

Decreasing the heat transfer coefficient to 100 W/m^2 K yields an improvement as shown in Figure 8, but the front still solidifies three quarters of the way up the tube.

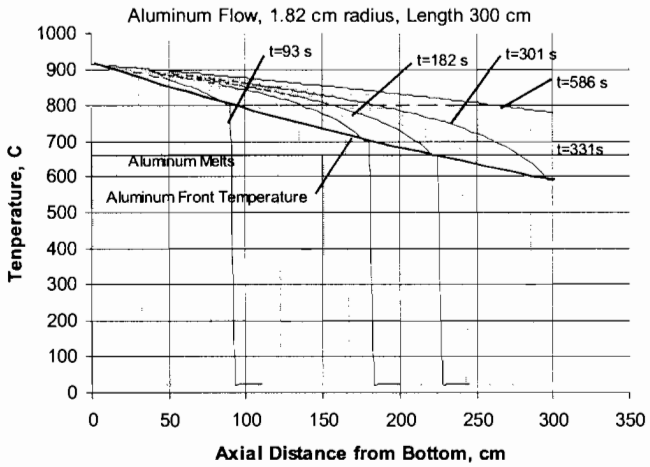


FIGURE 8 LOWER HEAT TRANSFER COEFFICIENT DELAYS SOLIDIFICATION.

Reduction of the heat transfer coefficient to 50 W/m^2 K yields the temperature profiles shown in Figure 9. The temperature reaches solidification at the tube exit so any heat transfer coefficient less than this will not plug up the tube. The minimum heat transfer coefficient for the liquid aluminum (which assumes pure conduction) can be estimated by setting the Nusselt number equal to two so that

$h = 2* k/D = 2*237\ W/mK/2*1.82\ cm = 13022\ W/m^2K.$

The minimum conductivity of the material to be used to construct a liner to resist the heat transfer between the aluminum and the CWF can be estimated by using the equation for series conductors and a wall thickness of 1 mm.

$$\frac{k}{th} = \frac{1}{\frac{1}{50} + \frac{1}{13022}} = 49.8 \frac{W}{m^2 K} \qquad k = 0.001m * 49.8 = 0.0498 \frac{W}{m K}$$

where k=the thermal conductivity of the insulator and th is the thickness.

This is a very low thermal conductivity. Fiberfrax, a ceramic tube, or some other insulator might be successfully used for this purpose. It should be noted that the actual value estimated for the effective aluminum heat transfer coefficient is small in determining the thermal conductivity of the insulator. So a heat transfer fluid with a lower k would do as well as aluminum.

It is also noted in Figure 9 that the temperature of the aluminum stream continues to increase after the fluid front leaves the CWF and eventually the entire aluminum temperature approaches 915 C.

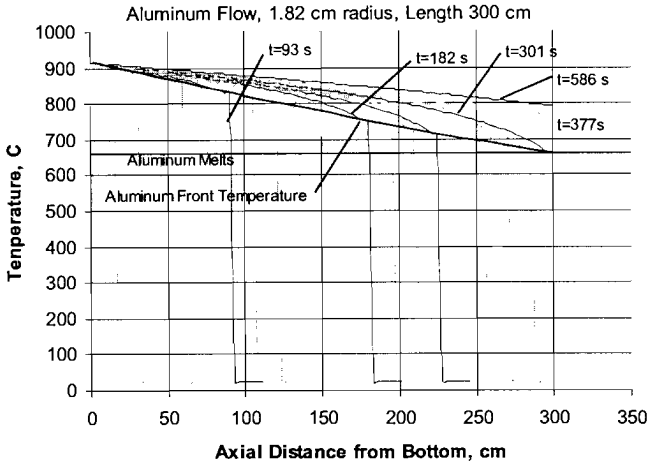


FIGURE 9 HEAT TRANSFER COEFFICIENT WHICH JUST REACHES SOLIDIFICATION.

The radial temperature profile for this case is shown in Figure 10. It appears from this figure that the temperature at the inner surface is almost at 915 C for the whole transient. In fact, the minimum temperature in the CWF at 40 hours is only 1.3 C less than that of the reference case. The heat up with the heat transfer resistance is almost the same as if there were no resistance the whole transient. Thus a material of smaller k could be used for heat up. However, aluminum is one of the few materials that has a relatively low solidification temperature (660 C) and a very high boiling point (2450 C).

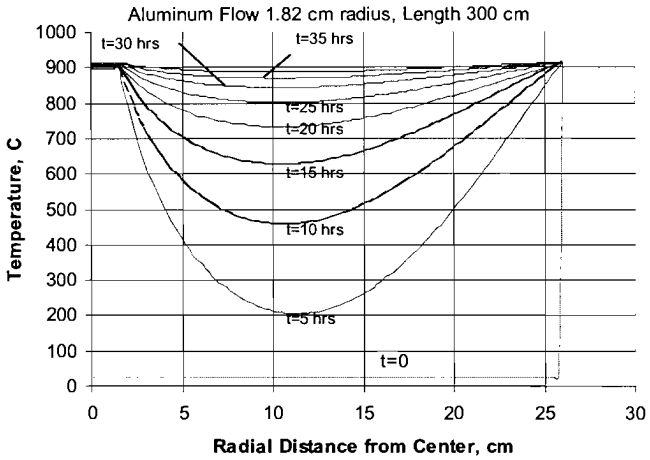


FIGURE 10 RADIAL TEMPERATURES FOR THE NON SOLIDIFICATION CASE.

So we conclude that as long as the center hole can be filled up with aluminum before it becomes plugged, the remainder of the heat up will be just as fast as the reference case. The heat transfer from the aluminum during the initial front traverse of the tube is the highest of the transient and is equivalent to passing the liquid aluminum over a 25 C CWF surface while the aluminum is cooling. Eventually it must solidify if the tube is long enough, so the insulating tube reduces the heat transfer.

One problem remains for the initial start up with this heat transfer coefficient of 50 W/m^2 K. That is the interface temperature between the aluminum and the CWF which is shown in Figure 11. The lower the heat transfer coefficient is between the metal and ceramic, the lower the interface temperature is. As is seen in Figure 11, the interface temperature quickly drops below solidification (less than 18 seconds) and remains there for over 586 seconds into the transient. This means that the aluminum near the wall solidifies and reduces the flow area to a small degree. Because the radial temperature gradient in the aluminum is so large, only a small amount solidifies. Eventually it melts again as the interface temperature makes it above the solidification temperature so it is not a long term problem. It does not effect the long term heatup.

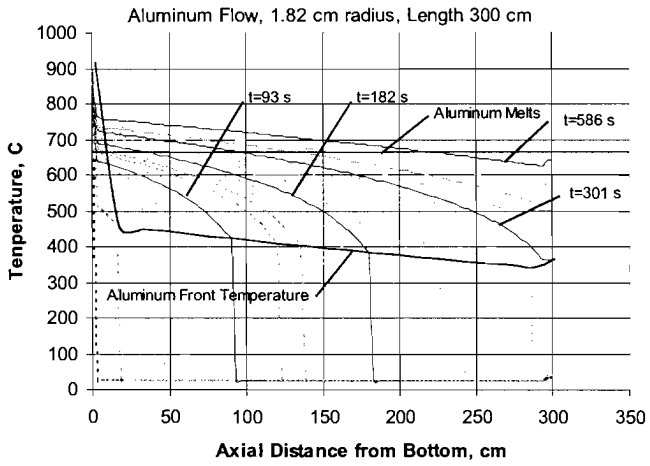


FIGURE 11 INTERFACE TEMPERATURE FOR H=50 W/M^2 K SHOWING SURFACE SOLIDIFICATION.

Another method of getting the initial aluminum out of the tube before it solidifies is to increase its velocity to reduce the residence time to the point where the aluminum exits the end of the CWF as a liquid. Figure 12 shows the result of increasing the velocity to 2.5 cm/sec. It is seen that the exit temperature in this case is 700 C so that this represents almost the minimum velocity that can be used without heat transfer resistance.

In this case since the heat transfer coefficient is very high, the interface temperature is close to the aluminum temperature so that no solidification of the aluminum would occur. The long term temperature profiles are very close to the reference case as well as the radial profiles. Although this velocity is needed for the time it takes the aluminum to fill the tube and bring the interface temperature above solidification, the velocity can then be decreased back to 1 cm/sec for the rest of the transient. Switching flow directions back and forth as initially envisioned at the beginning of this section will work since the inner wall (interface) temperature will remain over 900 C. Increasing the velocity is the preferred method of getting the aluminum front through the ceramic.

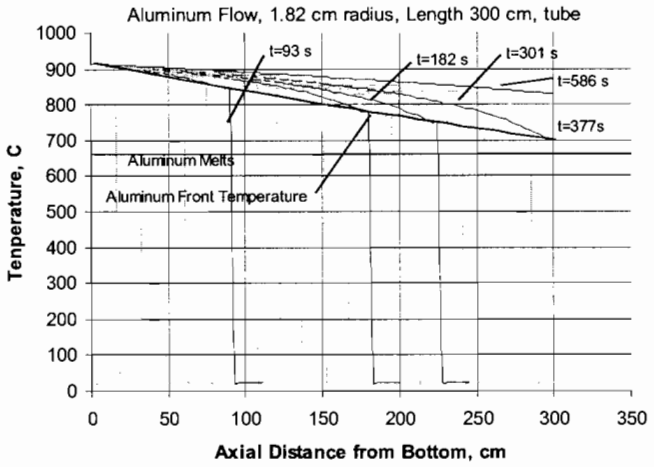


FIGURE 12 RESULTS FROM INCREASING THE ALUMINUM VELOCITY TO 2.5 cm/sec.

One final computational note in this section. The minimum CWF temperature no longer occurs on the axial midplane because the problem is non symmetric. However, in all the above long term graphs, the minimum temperature is plotted so that when that temperature exceeds 900 C, the entire CWF is above 900 C. Also, since the aluminum supply temperature is at 915 C, none of the CWF reaches temperatures above this.

Flowing a gas through the center hole

The consolidation of the CWF takes place in an argon gas-filled cell. Thus if a gas is to be used as a heat transfer medium, using argon to heat the center hole would make the problem of flowing gas through the hole simpler physically since there would be minimal concerns about degrading the atmosphere in the cell from leakage. For example, the concentration of oxygen in the cell is rigidly controlled to less than 50 ppm to prevent any metal oxidation.

In this section, we investigate the improvements that could be obtained with argon. This is a scoping study and is intended to estimate how well argon could perform in heating the ceramic. The same equations are used as for the aluminum case with the density and specific heat of air used instead of aluminum. Even though the density and velocity of the gas varies as it flows through the tube, the energy equation derived in Section 2 is still correct because the continuity equation allows ρ to be taken out of the time derivative and $\rho * V$ to be taken out of the axial derivative. Even though ρ and V are not constant along the tube, it will be assumed so for this scoping analysis. Also, the properties of air are used rather than that of argon realizing that air will produce somewhat better heating than argon. The effect of argon properties are then factored in.

The conditions necessary for adequate heat transfer with air can be estimated from the solution for aluminum from the previous section. The energy brought into the CWF in the aluminum flowing through the tube for the first run (in the previous section) heated up the CWF in 43 hours (Figures 5 and 6). This run may be used since there is not solidification concern for air.

$$\rho_m C_{Pm} V_m$$
$$= 2.7 gm/cc^3 * 0.903 J/gK * 1.0 cm/\sec$$
$$= 2.44 W/cm^2 K$$

The velocity needed by air at atmospheric pressure to bring in that much energy is given by the ratio of this product for the metal and air. Solving for this velocity yields

$$V_a = \rho_m C_{Pm} V_m / \rho_a C_{Pa}$$
$$= 2.44\ W/cm^2\ K / (0.000285\ g/cc * 1.01\ J/g\ K)$$
$$= 8463\ cm/\sec$$

(Note, the density of air corresponds to atmospheric pressure and 915 C). The same energy input could be attained by increasing the air density (and pressure) by a factor of 100 and decreasing the velocity by 100. In fact, the two solutions will be approximately the same if the transient term in the fluid energy equation is small in comparison to the axial term. The time step required for accuracy and stability for large velocities is approximately $\Delta z/V$. This is much smaller than that required for the CWF. Hence, it requires many time steps to get anything to happen in the CWF. Although an implicit axial derivative can be used in the gas energy equation to maintain numerical stability, the accuracy of the solution deteriorates for time steps much larger than $\Delta z/V$. Hence, the small time step required for accuracy was used.

To minimize computational time, a solution was obtained for the largest practical pressure (195 P_{atm}) to obtain the smallest velocity that could be used for a solution. Then, this solution can be used to determine the conditions necessary for obtaining adequate heat transfer with argon at different densities and velocities. In order to test out the above hypothesis, i.e., that the same input energy for the heat transfer fluid will yield the same CWF heatup rates, several runs were made out to 1 hours of heat up time. The first was a rerun of aluminum with the axial conduction term restored. The second and the third runs kept the axial conduction and increased the velocity and decreased the pressure to keep the product of velocity and density the same. The results included in Table 1 show that the minimum temperature of the fluid is basically the same at one hour. Therefore, the inner surface area temperature will be the same in all three runs so the heating time will be the same for the long term. That is, the results in Figures 5 and 6 apply to all three runs, not just the first. The third run was just about at the limit for the computer used since the time step was 1.66×10^{-5} hours. Fortunately the density was close to a high pressure air case. Consequently, this shows that the same input energy will yield approximately the same CWF heat up rate.

Table 1 ILLUSTRATION OF SIMILAR RESULTS FOR RUNS WITH THE SAME ENERGY INPUT.

	Density, g/cc	Velocity, cm/sec	Midplane Gas Temperature at 1 hour, C
ρ_{alum}	2.702	1	884.5
$0.1*\rho_{alum}$	0.270	10	884.8
$0.02*\rho_{alum}$	0.054	50	884.8
Air 195 atm	0.055	50	888.7

The last case in Table 1 is for air at a pressure of 195 atmospheres and 915 C. The calculated temperature is somewhat higher than the other cases because the heat capacity used is higher per unit mass for air than for aluminum (1009 J/kg K for air versus 903 J/kg K for aluminum). Nonetheless, the air results show that air will help in heating as well as the aluminum would. Using the above conclusion, a lower pressure of 195/5 = 39 atmospheres air would work well as long as the velocity was increased to 250 cm/sec. Air at atmospheric pressure could be used as long as the velocity is increased to 195 x 50 = 9750 cm/sec. (Note, the speed of sound is over 30,000 cm/sec at room temperature, so 10,000 cm/sec would produce significant pressure drops so higher pressure, lower velocity may be more desirable.) These results would also apply to argon as long as $\rho_{ar}\ C_{p,ar}\ V_{ar}$ is approximately equal to 2.44 W/cm^2 K. The density of argon is about 1.37 times that of air and the specific heat about 0.52 so the argon velocity would have to be 1.42 times that of air.

All of the above results assumed a heat transfer coefficient of 50 W/m^2 K, which is an intermediate value. The minimum value (laminar) for aluminum was 13022 W/ m^2 K and is far larger than the required 50 W/m^2 K. The heat transfer coefficient for air was estimated from the Dittus Boelter correlation. It may over estimate the value of the heat transfer coefficient because the Reynolds Number is estimated to be 14.6, clearly laminar flow. (Note, the product of density and velocity are the same in these three cases so the Reynolds number does not change if the viscosity does not change with temperature.) However, if one uses this correlation, the Nusselt number is 7.48 and the heat transfer coefficient is 55.5 W/m^2 K which meets the requirement. In fact, some turbulence or heat transfer enhancer may be required to obtain a high enough heat transfer coefficient, which could be part of the heater required to heat the gas to 915 C or fins in the tube. The thermal conductivity of argon is 66% that of air so additional augmentation of the heat transfer coefficient would be needed for argon over that of air. Future experiments may be used to determine the adequacy of the heat transfer coefficient.

Inserting a rod heater in the center hole

The purpose of this section is to determine the necessary rating of the heater rod that will be inserted into the CWF center hole to aid in the heatup. A heater rod would be turned on at its full rating and left on until the target temperature of 915 C was reached. After that, a thermocouple and a control circuit would be used to turn the heater off if the surface midplane temperature exceeded 915 C and turn it on if the temperature dropped below that. A heater power of 100 W was found to be somewhat too small. With this power, the CWF minimum temperature reached only 832 C in 40 hours compared to 891 C for the reference. The 500 W heater caused the minimum temperature to reach 883 C in 40 hours and the 1000 W heater brought the minimum temperature up to 891 C, which is almost as good as the reference.

The temperature profiles in the midplane for the 1000 W heater are shown in Figure 13. The interface temperature on the heater is seen to increase to the object temperature in approximately 8 hours. Even though this temperature is not at 915 C for the whole transient as it was in the reference case, the minimum CWF temperature is nearly as high as the reference case at 40 hours. Some temperature oscillation is observed once the heater has to turn on and off. This is caused by controlling on the midplane temperature. The upper and lower sections of the CWF reach temperatures higher than 915 C before the heater is turned off. These temperatures are no longer restricted to 915 C because the source is a rod of uniform axial heat generation. In practice, it may be better to use an axially shaped heat generation rate.

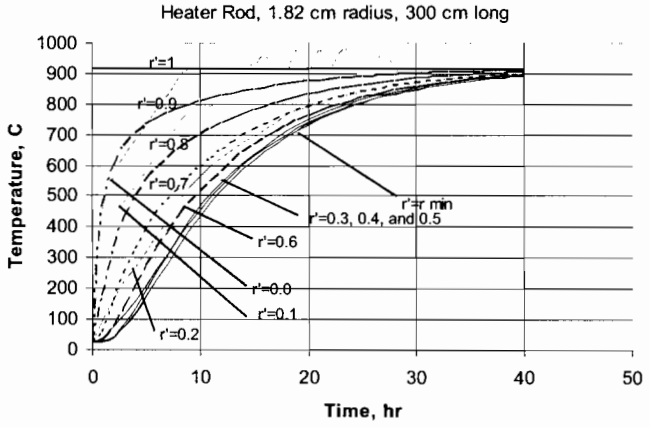


FIGURE 13 CWF TEMPERATURES OBTAINED WITH A 1000 W HEATER.

The conclusion of this section is that a 500 W heater is adequate to significantly decrease the heating time required to reach the object temperature. A 1000 W heater would improve the heating a bit more but not significantly. A 100 W heater is too small.

CONCLUSIONS

Several methods have been investigated to improve the heat transfer to the ceramic waste form that results from pyroprocessing spent nuclear fuel from the EBR II reactor. All have concentrated on options using center hole heat transfer to reduce the formation times. The CWF densifies during the heating and it is required to maintain a right cylinder shape during this consolidation. The material can consolidate unevenly and form a dome-shaped top if left without something to prevent this. The surface is kept flat by using a heavy plate on the top of the CWF which applies pressure throughout the

consolidation. Multiple intrusions into the CWF would make it difficult for this compression to work. It is possible, however, to use a single center hole and still allow the plate to compress the CWF. This is the reason limiting this investigation to center hole heat transfer for production runs.

The first method, simply pouring high temperature liquid aluminum into the hole, did not produce any noticeable affect on reducing heat up times. The second method, flowing liquid aluminum through the hole, works well as long as the velocity is high enough (2.5 cm/sec) to prevent solidification of the aluminum during the initial front movement of the aluminum into the center hole. The velocity can be reduced to 1 cm/sec after the initial front has traversed the ceramic. This procedure reduces the formation time to close to that of the reference case (43 hours). The third method, flowing a gas through the center hole, works well as long as the thermal capacity times the velocity of the gas is equivalent to that of the flowing aluminum and the velocity is high enough to produce a high heat transfer coefficient. The fourth method, using an electric heater, works well. Heater sizes between 500 to 1000 Watts are adequate.

Any one of the latter three methods can be successful. Selection of one of these will depend on which method is used to assist in cooling the consolidated CWF from 900 C to a temperature at which it can be handled. Switching from a hot gas to heatup to a cold gas to cool down the CWF does seem to be the most straight forward solution if the heat transfer coefficient is or can be made adequate. These analytical studies are very useful in significantly reducing the experimentation required to develop a successful method of augmenting the heatup.

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