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## Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

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**To cite this Article** Solbrig, C. W. and Vaden, DeeEarl(2006) 'Electrorefiner Liquid Cadmium Cathode Crucible Thermal Shock', *Separation Science and Technology*, 41: 10, 1985 — 2001

**To link to this Article:** DOI: 10.1080/01496390600745750

URL: <http://dx.doi.org/10.1080/01496390600745750>

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## Electrorefiner Liquid Cadmium Cathode Crucible Thermal Shock

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**Abstract:** A liquid cadmium cathode is used in an electrorefiner to remove plutonium and minor actinides from spent nuclear fuel by pyroprocessing. Liquid cadmium in a beryllia crucible, originally at 35°C, is lowered into 500°C salt electrolyte to begin reprocessing. Crucible cracking from thermal stress would release cadmium into the liquid salt causing electrorefiner failure. This study's purpose was to predict if the ceramic crucible would fail. A handbook method showed it would. An analytical model eliminating two large conservatisms predicted no failure. A beryllia crucible preheated to 321°C was successfully immersed in electrorefiner salt without failure. The conclusion is that handbook methods can be severely conservative in predicting thermal stress failures for immersion in low thermal conductivity fluids.

**Keywords:** Pyrochemical processing, electrorefiner, thermal stress, vessel failure

### 1. INTRODUCTION

Spent fuel reprocessing is an important step in closing the fuel cycle to produce fuel for fast reactors which eventually utilizes use of all the uranium-238. The Argonne National Laboratory developed a method of fuel reprocessing (1) which uses high temperature electrochemical pyroprocessing to extract a mixture of plutonium and minor actinides from the spent nuclear metallic fuel from the Experimental Breeder Reactor (EBR-II).

Received 23 October 2005, Accepted 29 March 2006

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The purpose of this study is to make certain that the liquid cadmium cathode (LCC) needed to perform this separation in an electrorefiner (the Argonne Mark V electrorefiner) will not fail from thermal shock. The electrorefiner, for purposes here, may be viewed simply as a box with an upper gas region (argon gas) and a lower molten salt region. The LCC (liquid cadmium contained in a beryllia crucible) originally at the hot cell temperature ( $\sim 35^\circ\text{C}$ ) is brought into the gas space to slowly preheat it, and then is quickly immersed in the  $500^\circ\text{C}$  electrorefiner salt electrolyte. If a crucible were to fail during immersion, cadmium would be lost into the salt and cause electrorefiner failure. Having to repair the electrorefiner and replace the crucible would be costly and cause a significant delay due to specialty manufacturing (6 months). An added purpose here is to determine if less expensive crucible materials could be used.

The literature (2) indicates that for a ceramic, beryllia has a good tolerance for thermal shock because of its relatively large thermal conductivity. However, this reference indicates that, in this instance, the temperature difference between the salt electrolyte and the crucible is large enough to cause failure. The LCC is brought into the argon gas space which preheats it slowly (about an hour) to an intermediate temperature before submersion in the molten salt. As the crucible is lowered into the salt, the flat eight inch diameter base comes in first contact with the high temperature salt making the base the region with the highest thermal stress. The crucible is frustum shaped, larger diameter on top, walls sloping at an angle of 7 degrees, 4.75 inches high, and walls and base 0.5 inches thick.

The thermal stress acts to pull the upper part of the base apart because the bottom surface expands due to increasing temperature and the upper surface resists the expansion because of its lower temperature. Thus the base bottom half is in compression and the upper half is in tension. Tensile failure is of concern because of the low tensile stress ability of ceramics. This study was planned to determine if the straightforward handbook analysis would be adequate to assess the thermal stress or whether more detailed analysis would be required to produce a realistic evaluation.

The following sections document this study which proceeds from simplest to more detailed analyses.

The first uses a published method to estimate the thermal stress.

The second investigates a single region analytical solution which takes into account the vertical temperature distribution in the vessel base but assumes cadmium has the same thermal properties as the crucible and the bottom surface is subjected to the  $500^\circ\text{C}$  salt temperature instantaneously upon contact.

The third includes a two region numerical solution which takes the cadmium properties into account.

The fourth models the salt properties as well.

In the fifth, the additional purpose of using a less expensive material (alumina) was investigated.

The sixth section described confirmatory experimental results followed by conclusions.

## 2. THERMAL STRESS-HANDBOOK METHOD

The thermal stress in the crucible base is caused by the temperature distribution. Each plane in the base is at a different temperature and each has a different stress free length corresponding to that temperature. Since the planes are all intrinsically attached, the overall length of the plate is determined by the average temperature of the plate. The stress is calculated by assuming that each plane is forced to be the same length as the average length. The planes at a higher temperature than the average are, therefore, under compression and the planes at a lower temperature are in tension. The amount that plane  $i$  must be elongated to reach the overall length,  $\Delta L_i$ , is

$$\Delta L_i = L\alpha(T_{avg} - T_i)$$

where  $L$  = length of the plate determined by the average temperature,  $\alpha$  = thermal expansion coefficient,  $T_i$  = temperature of plane  $i$ , and  $T_{avg}$  = spatial averaged temperature of plate.

The tensile stress that is induced in a plane elongated by the length  $\Delta L_i$  is approximately

$$\sigma_i = \frac{E}{1 - \mu} \frac{\Delta L_i}{L}$$

where  $E$  = Young's Modulus,  $\alpha$  = Thermal Expansion Coefficient, and  $\mu$  = Poisson's ratio.

Substituting in for  $\Delta L_i$  yields the thermal stress,  $\sigma$ , at any plane,  $i$ , as

$$\sigma_i = \frac{E\alpha}{1 - \mu} (T_{avg} - T_i) \quad (1)$$

so the thermal stress,  $\sigma_{max}$ , on the surface (which is the maximum) can be calculated by

$$\sigma_{max} = \frac{E\alpha}{1 - \mu} (T_{avg} - T_{surf}) \quad (2)$$

where  $T_{avg}$  = Average temperature in the crucible wall and  $T_{surf}$  = Surface temperature

Substituting in the properties (3) for BeO,  $E = 50 \times 10^6$  psi,  $\alpha = 8 \times 10^{-6}$  in/in °C, and  $\mu = 0.26$  yields for the stress temperature coefficient for BeO.

$$\frac{E\alpha}{1 - \mu} = \frac{50 * 8}{1 - 0.26} \frac{\text{psi}}{\text{°C}} = 540.54 \frac{\text{psi}}{\text{°C}} \quad (3)$$

Ceramics usually have a much smaller tensile stress limit than compressive so will usually fail in tension. The compressive strength of BeO is 225,000 psi which is over 10 times higher than the tensile strength of 20,000 psi.

The failure temperature difference (i.e., the average to surface temperature difference causing tensile stress failure) is obtained by solving for the difference from Equation 3 with the failure stress,  $\sigma_{tensile}$ , substituted in for the maximum

$$(T_{avg} - T_{surf})_{fail} = \frac{\sigma_{tensile} * (1 - \mu)}{E\alpha} \quad (4)$$

which is the tensile stress limit divided by the stress temperature difference coefficient.

Dividing the stress temperature coefficient for BeO into its tensile strength, 20,000 psi, yields the BeO failure temperature difference

$$(T_{avg} - T_{surf})_{fail} = 20,000/540.54 = 37^\circ\text{C} \quad (5)$$

Assuming a linear temperature throughout the crucible wall would make the inside to outside failure temperature difference =  $2 \times 37^\circ\text{C} = 74^\circ\text{C}$ . Thus the crucible would have to be preheated to  $426^\circ\text{C}$  to stay below the thermal stress limit. This is undesirable due to the vapor pressure of cadmium at that temperature and long time (estimated at 3 hours) required to preheat the crucible to this temperature. The large vapor pressure would cause significant vaporization and transport of cadmium to the vessel walls in this time. Insertion of the crucible at or near the melting point of cadmium ( $321^\circ\text{C}$ ) is required to prevent this migration. Once the crucible is submerged, heating can continue without cadmium migration which is prevented by the salt covering.

As will be seen, this estimate is very conservative. The next section shows the temperature profile is not linear allowing the outer minus the inner temperature difference at failure to be larger than  $74^\circ\text{C}$ . The *Concise Encyclopedia of Advanced Ceramic Materials* (2) uses the failure temperature difference (Equation 4) to rate various materials for thermal shock resistance. It suggests the resistance of alumina is better than BeO so will be considered later in this study as an alternate material for crucible material.

### 3. THERMAL STRESS-ANALYTICAL SINGLE REGION SOLUTION

The vertical temperature profile in the crucible base was first estimated using the simplest analytical solution for transient heat conduction in a slab. In order to use a single region analytical solution, a constant BeO bottom surface temperature must be assumed equal to the salt temperature ( $500^\circ\text{C}$ ). The slab is 4

inch thick. The bottom 0.5 inches represents the BeO base and the top 3.5 inches represents cadmium. The upper surface of the cadmium is assumed insulated. The upper insulated surface at  $X = L$  implies two things. The first is that the heat transfer to the gas (during insertion) above the crucible is small. The second is that different initial depths ( $\pm 1$  inch) of the cadmium would not change the temperature distribution in the base significantly so the conclusions about the stress would apply to different cadmium depths. To obtain an analytical solution, as an approximation, BeO thermal properties were used for both the BeO and the Cd regions. This assumption is investigated later and shown to be somewhat non conservative.

The temperature solution (4) as modified for a slab of height  $L$  of initial temperature  $T_{init}$ , with an insulated upper surface ( $X = L$ ), and with bottom surface ( $X = 0$ ) subjected to temperature  $T_{salt}$  at time zero and thereafter is

$$\frac{T - T_{init}}{T_{salt} - T_{init}} = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)} \left[ \text{Exp} \left\{ -\alpha_t (2n+1)^2 \frac{\pi^2 t}{4L^2} \right\} \right] \cdot \cos \frac{(2n+1)\pi(X-L)}{2L} \quad (6)$$

where  $\alpha_t$  = thermal diffusivity =  $k/\rho C_p$ ,  $t$  = time,  $x$  = distance,  $k$  = thermal conductivity, and  $\rho$  = density,  $C_p$  = Specific heat.

The temperature profile in the base calculated assuming that the crucible is preheated to 300°C before immersing in the 500°C salt is shown in Fig. 1.

The cadmium is assumed to be liquid or soft enough (MP = 321°C) so it does not induce any stress into the BeO base. A non-zero thermal gradient is noted at  $X = 0.5$  inches. It is zero on the upper surface of the cadmium.

The above temperature distribution was used to calculate the thermal stress with Equation 1. The results are shown below in Fig. 2. Tensile stresses (i.e., positive stresses) are observed from the top surface of the BeO base down to about half the base thickness for some period of time. The

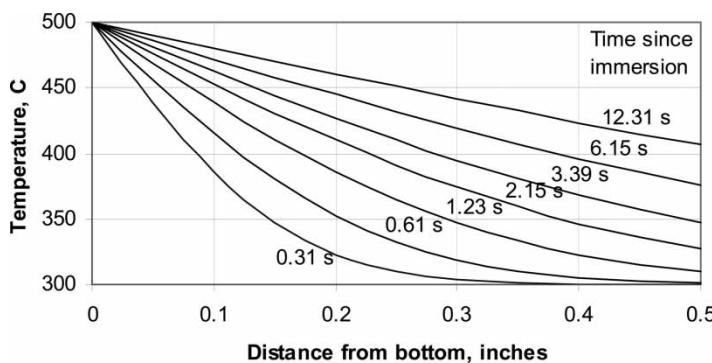


Figure 1. Spatial temperature distribution in the crucible base.

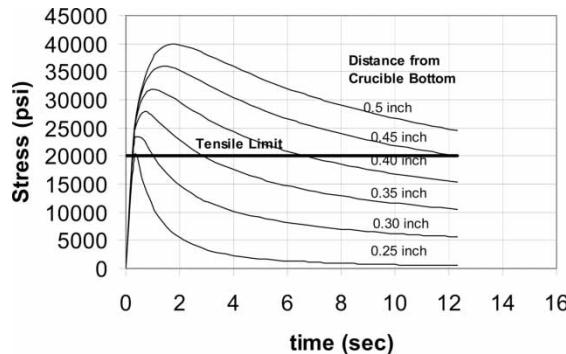


Figure 2. Stress in the base of the 300°C crucible immersed in 500°C salt.

stresses are largest on the top surface of the base and exceed the limit tensile stress of 20,000 psi for approximately 16 seconds. The stresses decrease with elevation. The maximum stress occurs at about 2 seconds. Even though this stress occurs for a very short time, experience with thermal shock in alumina posts immersed in liquid metal, broke catastrophically in a very short time, on the order of seconds. In this case, the top 20% of the base is above the tensile stress limit for over 6 seconds and this implies failure in this region which would lead to failure of the base. Therefore, based on this analysis, damage to the crucible due to thermal shock would occur. The lower part of the base is in compression with stresses reaching over 80,000 psi which is still much less than the compressive limit of 225,000 psi. The analytical temperature profile causes the lower surface compression to be higher than the straight line method and the tensile stress at the upper surface is less.

Dimensional analysis shows that all the stresses are proportional to the initial temperature difference between the salt and crucible. Thus decreasing this temperature difference by 2 will reduce the maximum stress from 40,000 psi to 20,000 psi. The thermal stress calculated for a 400°C crucible immersed in a 500°C bath produces the stresses shown in Figure 3 which are all below the tensile stress limit. The crucible would probably remain intact for this temperature difference. However, the vapor pressure of cadmium with subsequent possible migration would again be too high.

The thermal properties used in the above calculations (3, 5) for BeO are  $k_b = 50 \text{ W/m K}$ ,  $\rho_b = 2.85 \text{ gm/cc}$ , and  $C_{pb} = 0.25 \text{ cal/gm K}$ . The analysis assumes that the thermal conductivity is constant but it, in fact, changes significantly with temperature. A plot of the thermal conductivity for BeO obtained by two different methods (5) is shown (solid lines) in Fig. 4. A significant extrapolation is involved in using values up to 500°C. A straight line extrapolation (dashed lines) to 300°C yields a value of about 50 W/m C, the selection made for in the previous calculations.

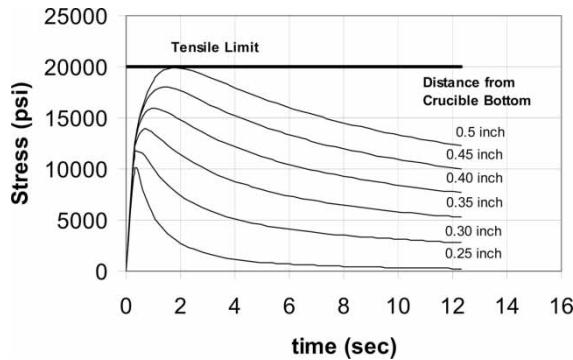


Figure 3. Stress in the base of the 400°C crucible immersed in 500°C salt.

Due to the uncertainty in this value, the effect of thermal conductivity is investigated in the following. In Fig. 5, the thermal conductivity was decreased by a factor of 2 to 25 W/m K. Clearly, the stress stays above the limit for a longer time. The surface stress eventually decreases below the tensile stress limit in 32 seconds.

Dimensional analysis shows that the only two independent parameters involved in the analytical solution is the above mentioned dimensionless temperature,  $\bar{T}$ , and the dimensionless time,  $\bar{t}$ , given by the equation:

$$\bar{t} = \frac{\alpha * t}{L^2} = \frac{k}{\rho \cdot C_p} * \frac{t}{L^2} \quad (7)$$

Thus, the temperature is not changed with a change in the thermal conductivity, and hence, from Equation 1, neither is the stress. Only the time at which it occurs is changed. If the thermal conductivity is decreased by a factor of two, then the time scale is lengthened by a factor of two as shown in Fig. 5.

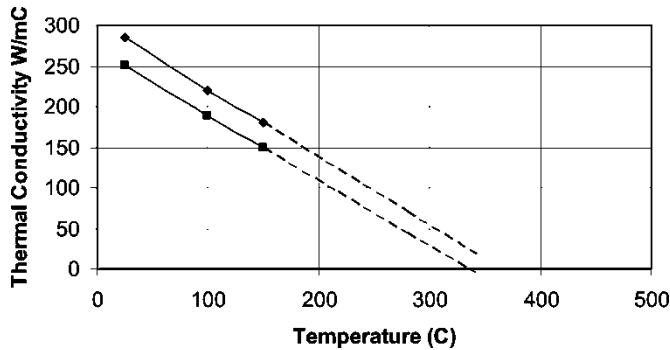
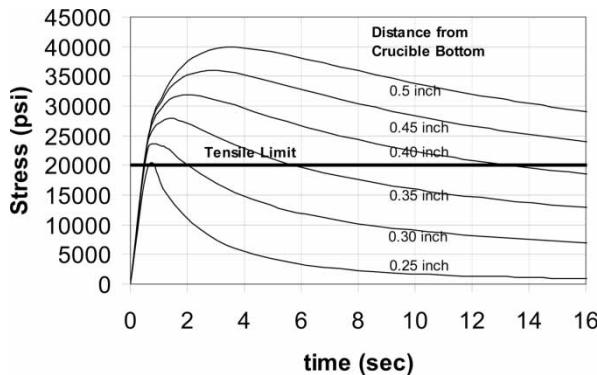


Figure 4. Thermal conductivity of BeO.



**Figure 5.** Thermal stress for a material with half the thermal conductivity.

According to Equation 7, the effect of increasing thermal conductivity by a factor of four from 50 to 200 W/m°C (the latter represents the low temperature conductivity) is to significantly reduce the time that the stress is above the limit. The stress level is not reduced, just the time at stress. The time when the maximum stress decreases below the tensile stress limit is reduced by a factor of 4 to just 4 seconds. The base may be able to withstand this stress level for such a short time. However, from the data presented in Fig. 4, it seems unlikely the thermal conductivity would remain as high as the low temperature value when in the 300°C to 500°C range.

As mentioned previously, Reference 2 uses Equation 4 to rate materials for thermal shock resistance. The author indicates this quantity should be used when the thermal shock occurs rapidly. Another parameter (the product of the material thermal conductivity and Equation 4) is to apply for slower thermal shock problems, however, a justification for such a term is not indicated. The above analysis shows that the stress levels do not decrease with increasing thermal conductivity, but the time that the stress must be endured is decreased so the material may be able to absorb the stress energy over a shorter time without failing giving some justification for this second parameter. The fast thermal shock parameter says alumina is better than BeO whereas the slow thermal shock parameter says that BeO is superior. The analysis in a latter section contradicts this later conclusion and shows alumina is also better for the slow thermal shock.

#### 4. THERMAL STRESS-TWO REGION SOLUTION WITH Cd PROPERTIES

Since the maximum stress occurs at the interface between the BeO and cadmium, any error in calculating the temperature profile in this location

could cause an error in the stress. The properties of cadmium are significantly different than BeO so a numerical solution was developed to allow the material properties to be changed easily at the interface. The numerical solution using BeO properties for both regions agreed with the analytical solution of the previous section. The properties (6) used for cadmium (Melting Point = 321.18°C) are  $k = 96.8 \text{ W/m K}$ ,  $\rho = 8.65 \text{ gm/cc}$  at 300 K, and  $C_p = 0.966 \text{ cal/gm K}$ . These compare to the BeO properties of  $k = 50 \text{ W/m K}$ ,  $\rho = 2.85 \text{ gm/cc}$ , and  $C_p = 0.25 \text{ cal/gm K}$ . Thus, although the thermal conductivity of cadmium is about twice that of BeO, the thermal capacitance,  $\rho C_p$ , is almost 12 times as high so it takes more energy to raise cadmium one degree than BeO. As will be seen, the large  $\rho C_p$  of the cadmium causes the temperature to stay low causing a large thermal stress.

The two boundary conditions between the two metals are

$$k_b \frac{\partial T_b}{\partial x} = k_c \frac{\partial T_c}{\partial x}; \quad T_b = T_c \quad (8)$$

Because the first boundary condition contains only the thermal conductivity and the partial differential equations contain the thermal diffusivity,  $\alpha_t = k/\rho C_p$ , the results depend on each of these two parameters independently and not just  $\alpha_t$  alone. Fig. 6 shows a slightly higher stress calculated in the crucible base with the Cd properties (46,000 vs. 40,000 psi) but the time above the tensile limit has increased considerably because the interface temperature stays lower than if the vessel were filled with BeO rather than Cd.

An expanded time period is presented in Fig. 7. It is seen that it takes over 280 seconds for the maximum stress to become less than the tensile limit so that the stress energy which must be absorbed by the beryllia is much greater than in the previous case.

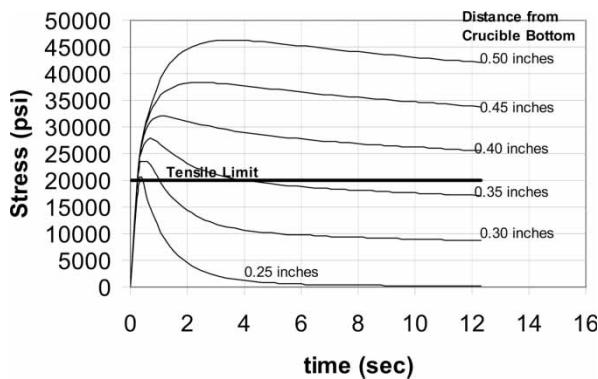


Figure 6. Stress calculated with cadmium modeled.

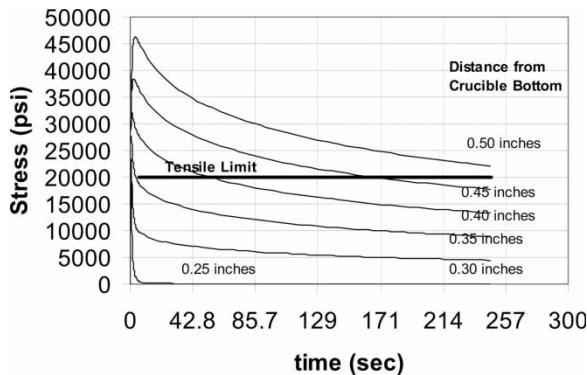


Figure 7. Stress calculated with cadmium modeled, long time.

## 5. THERMAL STRESS-TWO AND THREE REGION SOLUTION WITH SALT

Section 3 calculations assumed that the temperature of the salt was impressed instantaneously on the surface of the crucible base. In fact, the salt is a poor conductor and the heat transfer to the crucible is weak. A two region solution including BeO and the salt region was obtained. The salt region is modeled as a slab four inches thick directly under the crucible base. The salt is 44 wt% LiCl and 56 wt% KCl. The bottom surface of the salt slab is assumed constant at 500°C and the top surface is in contact with the bottom surface of the crucible base. The boundary heat transfer conditions, similar to Equation 8 of Section 4, to connect the two regions are

$$k_s \frac{\partial T_s}{\partial x} = k_b \frac{\partial T_b}{\partial x}; \quad T_s = T_b \quad (9)$$

The salt properties (7-8) at 500°C,  $k_s = 0.428 \text{ W/m K}$ ,  $\rho_s = 1.6225 \text{ gm/cc}$ ,  $C_{ps} = 0.2961 \text{ cal/gm K}$ , were used. Compare to  $k_b = 50 \text{ W/m K}$ ,  $\rho_b = 2.85 \text{ gm/cc}$ , and  $C_{pb} = 0.25 \text{ cal/gm K}$  for beryllia. The thermal conductivity of salt is 100 times less than that of beryllia. The temperature profile is shown in Fig. 8. Even though the salt is at 500°C, the bottom crucible base surface temperature has only increased from 300°C to less than 312°C in 12.3 seconds. This is significantly different than the 500°C salt temperature instantaneously assumed impressed on the surface in the previous analyses. The salt temperature at the interface matches that of the beryllia as specified in the second boundary condition in 9 and the first condition forces the interface temperature gradient in the salt to be 100 times greater than that in the crucible.

The maximum stress occurs in less than 6 seconds. With this slow surface temperature rise, the stresses are considerably lowered. The results are shown in Fig. 9. The maximum stress is less than 1800 psi, much less than the tensile

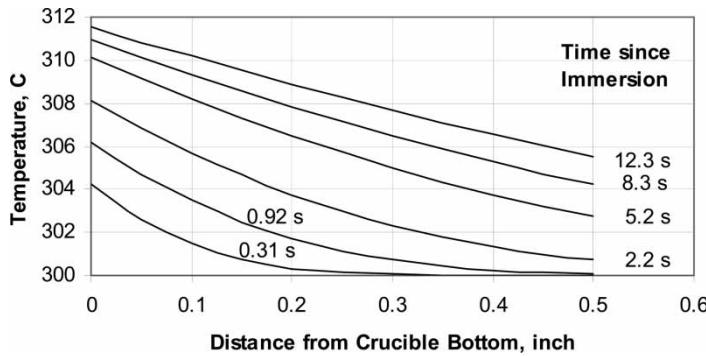


Figure 8. Temperature profile in the crucible base with salt modeled.

limit of 20,000 psi and a reduction by a factor of over 20. No crucible damage is expected.

Three additional factors were looked at in this section to determine the robustness of this conclusion. The first factor looked at was the resistance between the beryllia and the cadmium. Poor contact was simulated by assuming the top of the crucible base was insulated. The stresses are even lower in this case as shown in Fig. 10.

The second factor, including cadmium also, was investigated with a three region solution with salt, beryllia, and cadmium. The resultant stress is shown in Fig. 11. As observed in Section 4, the replacement does increase the calculated stress by about 30% but not enough to challenge the crucible integrity.

The final factor investigated is the possible increase of heat transfer due to circulation in the salt. Salt melts at around 352°C, so it is a liquid at 500°C. There is induced natural circulation in the electrorefiner due to the heat input from the bottom and side heaters on the vessel. There is also a salt

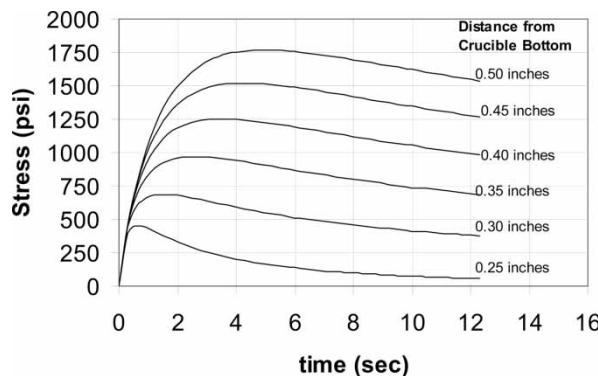
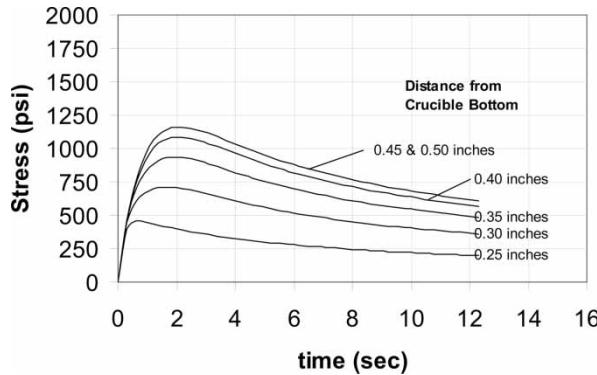


Figure 9. Stress profiles in the crucible base with salt modeled.

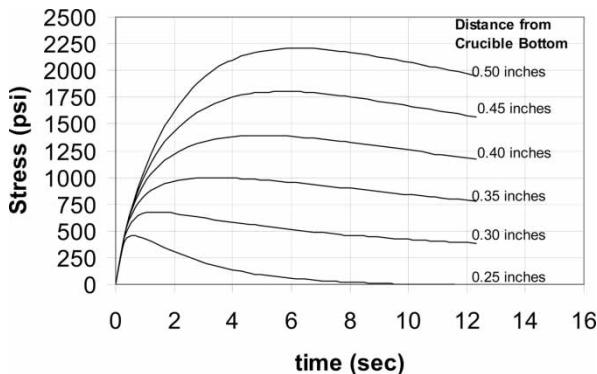


**Figure 10.** Stress calculated assuming poor beryllia cadmium contact.

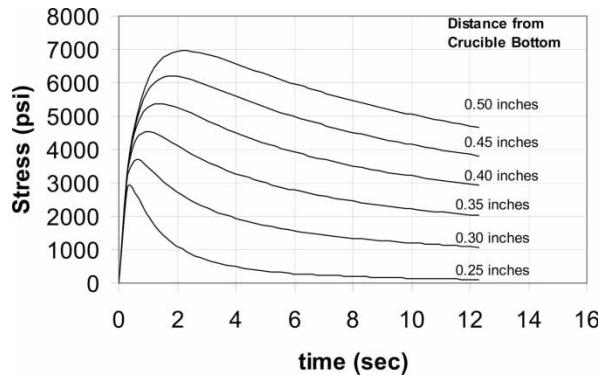
stirrer, so there is forced convection. Also, the cold crucible immersed in the upper region of the hot salt could cause circulation. To estimate the possible heat transfer effect of this convection, the salt thermal conductivity was increased by a factor of 10. The lower surface reached 337.3°C in 12.3 seconds versus 311.5°C for the stagnant salt. This higher temperature caused higher stresses as shown in Fig. 12.

The maximum stress is about 7000 psi which is almost four times that of the stagnant salt but still significantly below the tensile stress limit so it is not expected that the circulation would cause the crucible base to fail.

For this case, the temporal behavior of the crucible base surface temperatures as well as the average temperature is shown in Fig. 13. It is seen that the lower surface temperature increases quickly to 335°C due to large initial heat fluxes then exhibits a slower increase rate as the temperature profiles flatten out. The maximum temperature difference between the average and the



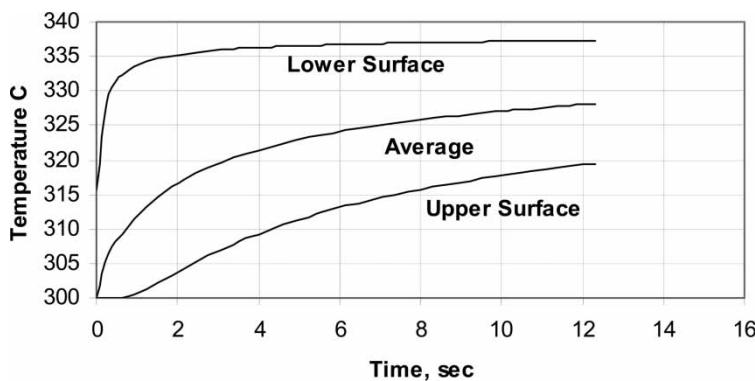
**Figure 11.** Stress calculated using cadmium properties in the cadmium region.



**Figure 12.** Stresses calculated assuming significant circulation in the salt.

upper surface temperatures occurs at 2.15 seconds and then decreases monotonically from that point on. Therefore, the maximum stress occurs at 2.15 seconds. Crucible heat up to 500°C takes about an hour.

Hence, the conclusion of this section is that the stress introduced into the crucible is small enough that no damage will occur. Assuming, as was done in the previous two analyses, that the salt temperature was instantly impressed on the surface of the crucible base is very conservative. In fact, the crucible could withstand insertion into the 500°C salt even if it were preheated to only 100°C. That is, dimensional analysis shows that the stress for a crucible inserted at 100°C may be obtained from any of the cases presented in this section by multiplying the 300°C insertion results by the ratio  $(500^\circ\text{C} - 100^\circ\text{C})/(500^\circ\text{C} - 300^\circ\text{C}) = 2$ . The resulting stress in all of the above cases in this section would still be significantly less than the limit stress of 20,000 psi.



**Figure 13.** Temperatures which determine the maximum stresses.

## 6. THERMAL STRESS-ALUMINA SALT RESULTS

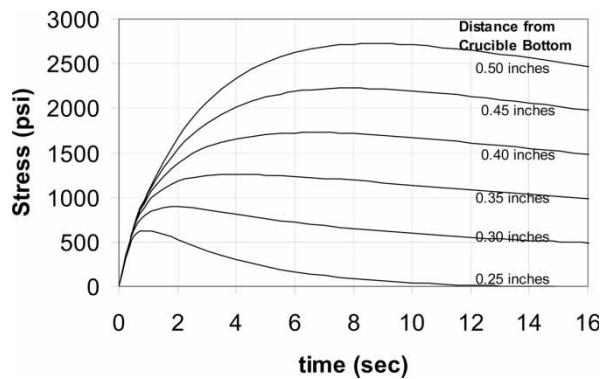
Due to the cost and time to fabricate beryllia crucibles, alumina was investigated as a possible alternate crucible material. It is desired to have multiple crucibles available to increase the electrorefiner throughput.

The properties of alumina were used in the two region program to determine the ability of alumina to withstand the thermal shock. The thermal properties (11) used were  $k = 35 \text{ W/m K}$ ,  $\rho = 3.89 \text{ gm/cc}$ ,  $C_p = 0.209 \text{ cal/gm K}$ . The structural properties (11) used were  $E = 54.4 \times 10^6 \text{ psi}$ ,  $\alpha = 8.4 \times 10^{-6} \text{ in/in } ^\circ\text{C}$ ,  $\mu = 0.22$ . The stress temperature coefficient for alumina (from Equation 3) is

$$\frac{E\alpha}{1 - \mu} = \frac{54.4 * 8.4 \text{ psi}}{1 - 0.22 \text{ } ^\circ\text{C}} = 585.85 \frac{\text{psi}}{^\circ\text{C}}$$

As with beryllia, alumina has a much smaller tensile stress limit than compressive but its tensile limit is much higher than beryllia. The compressive strength of  $\text{Al}_2\text{O}_3$  is 304,500 psi and the tensile limit is 50,000 psi. The tensile stress limit is 2.5 times that of beryllia. The tensile failure temperature difference from Equation 4 is  $50,000 \text{ psi} / 585.85 \text{ psi/}^\circ\text{C} = 85^\circ\text{C}$  which is much larger and less limiting than that of beryllia and is consistent with that reported in the literature (2).

Consistent with this conclusion, stress calculation show that alumina is able to withstand higher levels of thermal shock. The stress calculation is shown in Fig. 14. Although the calculated stress is higher (2700 psi) than in the corresponding BeO calculation (1800 psi), the limit stress for alumina is 50,000 psi which is significantly higher than the BeO stress limit (20,000 psi) so alumina is expected to survive this temperature service better than BeO.



**Figure 14.** Stress in a  $300^\circ\text{C}$  alumina crucible base immersed in  $500^\circ\text{C}$  salt.

This model also shows that a 100 C alumina crucibles will also survive immersion when immersed in the 500°C salt. The maximum stress reached is higher but less than 4700 psi, much lower than the 50,000 psi tensile stress limit.

Thus this less expensive material is satisfactory for future crucible construction of crucibles. This material is easier to obtain and easier to fabricate than beryllia.

## 7. EXPERIMENTAL CONFIRMATION

Based on the positive results in Section 5, the beryllia crucible loaded with a new cadmium ingot was inserted into the electrorefiner salt. The cadmium ingot, a right circular cylinder with rounded edges and of a smaller diameter than the crucible insides was loaded into the crucible and the combination inserted into the Mark V electrorefiner in the gas space above the molten salt for preheating. Because no temperature indicators existed in the cadmium or the beryllia crucible, the rod used to make electrical contact with the cadmium was used to determine if the cadmium had reached melting by lowering it into the cadmium with the motor torque set to stall if a solid cadmium surface was encountered. After four hours in the gas space, cadmium melting was detected so the crucible had reached the 321°C melting point of cadmium. The crucible was then lowered into the salt so that electrochemical transport could take place. The Mark V electrorefiner operated successfully for 28 hours to gather Pu/U from the salt indicating the crucible stayed intact for this time.

After the experiment was terminated, the crucible was raised to the cover gas space to cool overnight. Then it was removed from the electrorefiner. The solidified cadmium was removed from the crucible and both the solidified cadmium and crucible were examined for possible damage. The examination confirmed that no crucible failure occurred during the run. During the run, molten cadmium conformed to the frustum shape of the crucible. Cracks or crucible failure would have been mirrored in the solidified cadmium. No damage was observed in the cadmium.

The above theory predicts that the most severe thermal stress occurs on the inside of the crucible. Both the inside and outside of the crucible were examined but the inside received the most scrutiny. Although the crucible inside was discolored from contact with the cadmium no failure or cracks observed. Since none occurred, the conclusion reached in the paper is confirmed that no damage to the vessel would occur.

Since the first run, two more successful runs have been made with no damage to the crucible occurring during the electrorefining, solidification, or removal from the electrorefiner. In both these latter runs, the cadmium bonded to the BeO but this was not caused by thermal stress.

Although a full size alumina crucible has not been used yet, small alumina crucibles were used for five LCC tests in a lab scale electrorefiner. In four of the tests, the crucibles were held above the molten salt for an hour before transfer into the 500°C salt. None of these evidenced any cracks or failure. In the fifth test, the crucible was directly transferred from the hot cell temperature (~35 C) into the 500°C salt. Even in that case, there was no damage to the crucible (12).

## 8. CONCLUSIONS

A series of analytical models were used to investigate the possible thermal stress damage to a beryllia crucible preheated to 300°C and inserted into a 500°C electrolyte salt. Handbook formulas and an analytical solution assuming the salt temperature is impressed instantly on the surface of the crucible showed the crucible will fail. These analyses are far too conservative for immersion of objects in low thermal conductivity fluids. Analyses which take the low thermal conductivity of the salt into account show that the crucible will not fail under these conditions. Three electrorefiner runs have been made with beryllia crucibles which confirmed that damage does not occur due to this temperature excursion in the beryllia. Several small alumina crucibles also survived the same temperature cycle in a lab scale electrorefiner.

General conclusions can be drawn from the above analyses for solid objects being plunged into liquids at much higher temperatures. The simple solutions (handbook and one region analytical) which assume the bulk fluid temperature is immediately impressed on the surface of the solid apply only if the thermal conductivity of the fluid is high relative to that of the solid. In all other cases, they are much too conservative because the interfacial temperature differs from that of the fluid. This temperature will be closest to average temperature of the material with the highest thermal conductivity; so the interface temperature will be close to the fluid for high effective thermal conductivity fluids and close to the solid for low thermal conductivity fluids. The closer this interfacial temperature is to the initial solid temperature, the lower the tensile thermal stress will be. Since the highest thermal stress is on the inner surface, the conductivity of the fluid in the crucible also makes a difference with the stress increasing for a high thermal conductivity inner fluid and lasting a longer time.

Except in cases with a large conductivity fluid, it is necessary to perform a detailed analysis to obtain realistic results. In fact for very low fluid thermal conductivities (relative to the solid) it would be better to perform no analysis rather than a handbook one because the thermal stress is probably not a problem since the low conductivity fluid cannot transfer the heat to the solid. Care should be taken in attempting to apply these conclusions to a

hot crucible being quenched in a cold fluid because the tensional thermal stress would be on the outside (bottom) of the vessel rather than the inside.

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