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Glovebox Advanced Casting System Casting Optimization

Fuel Cycle Research & Development
Advanced Fuels Campaign

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U. S. Department of Energy
Office of Nuclear Energy

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ABSTRACT

Casting optimization in the GACS included three broad areas; casting of U-10Zr pins, incorporation of an integral FCCI barrier, and development of a permanent crucible coating. U-10Zr casting was improved over last year's results by modifying the crucible design to minimize contact with the colder mold. Through these modifications casting of a three pin batch was successful. Incorporation of an integral FCCI barrier also was optimized through furnace chamber pressure changes during the casting cycle to reduce gas pressures in the mold cavities which led to three full length pins being cast which incorporated FCCI barriers of three different thicknesses. Permanent crucible coatings were tested against a base case; 1500°C for 10 minutes in a U-20Pu-10Zr molten alloy. None of the candidate coating materials showed evidence of failure upon initial visual examination. In all areas of work a large amount of characterization will be needed to fully determine the effects of the optimization activities. The characterization activities and future work will occur next year.

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GACS Casting Optimization and Quality Assurance Assessment

1. INTRODUCTION

The Glovebox Advanced Casting System (GACS) was installed and initially operated in a minor actinide qualified glovebox in the summer of 2013. This furnace was designed to provide a flexible casting development platform where both gravity casting into a permanent mold and counter gravity casting into a glass mold could be developed and studied. In addition to casting development, the GACS will also be used to study melt properties and general sample fabrication.

The solid model and photograph of the GACS configured for gravity casting is shown in Figure 1. As can be seen in this figure, the GACS is contained in a stainless steel shell that uses standard ISO style vacuum flanges for all sealing surfaces. The mold and crucible are independently inductively heated. The induction power supply is sized large enough to allow rapid heating of the crucible in order to decrease the total amount of time the fuel alloy is heated, thereby reducing volatility losses. A number of thermocouples can be monitored throughout the heating cycles. A standard gravity casting utilizes four K-type thermocouples along the length of the mold with an option of monitoring up to six additional K-type thermocouples. A total of four B-type thermocouples can be used as well: two thermocouples are used on the crucible, one is used to monitor the crucible induction coil temperature, and one thermocouple is a spare that can be used where needed. A total of five pressure transducers can be used to monitor the furnace system, although currently only two are being utilized in the gravity pour configuration. The current design uses a graphite crucible and a two-piece graphite mold with the pins being cast on the parting line of the mold.

During the FY14 casting campaign only limited success was seen. The main alloy focused on was U-10wt% zirconium (U-10Zr) however, the zirconium was not fully incorporated into the melt. Despite this set back various mold and process improvements were made which were verified by the appearance of the uranium alloy with some amount of zirconium that was cast, although the bulk of the zirconium remained in the crucible. At the end of FY14 to investigate the inability to dissolve zirconium a charge of pure copper was done in which a thermocouple was placed in the standard location on the outside of the crucible and one was place in the crucible, just contacting the molten alloy. From this experiment it was seen that melt temperature was >100° C lower than the measured temperature on the crucible. During FY15 casting development continued to use U-10Zr alloys, although some plutonium bearing charges were also used. Work was focused on overcoming the >100° melt-crucible differential which leads to zirconium dissolution problems, casting into integral fuel cladding chemical interaction (FCCI) barriers, and testing of candidate crucible coating materials. The crucible coating studies were done in collaboration with the Korean Atomic Energy Research Institute. Coating are of interest to allow re-use of the crucibles without additional coating and to mitigate any possible interaction with fuel components, including contaminates especially in a simulated recycled fuel composition, and the graphite crucible.

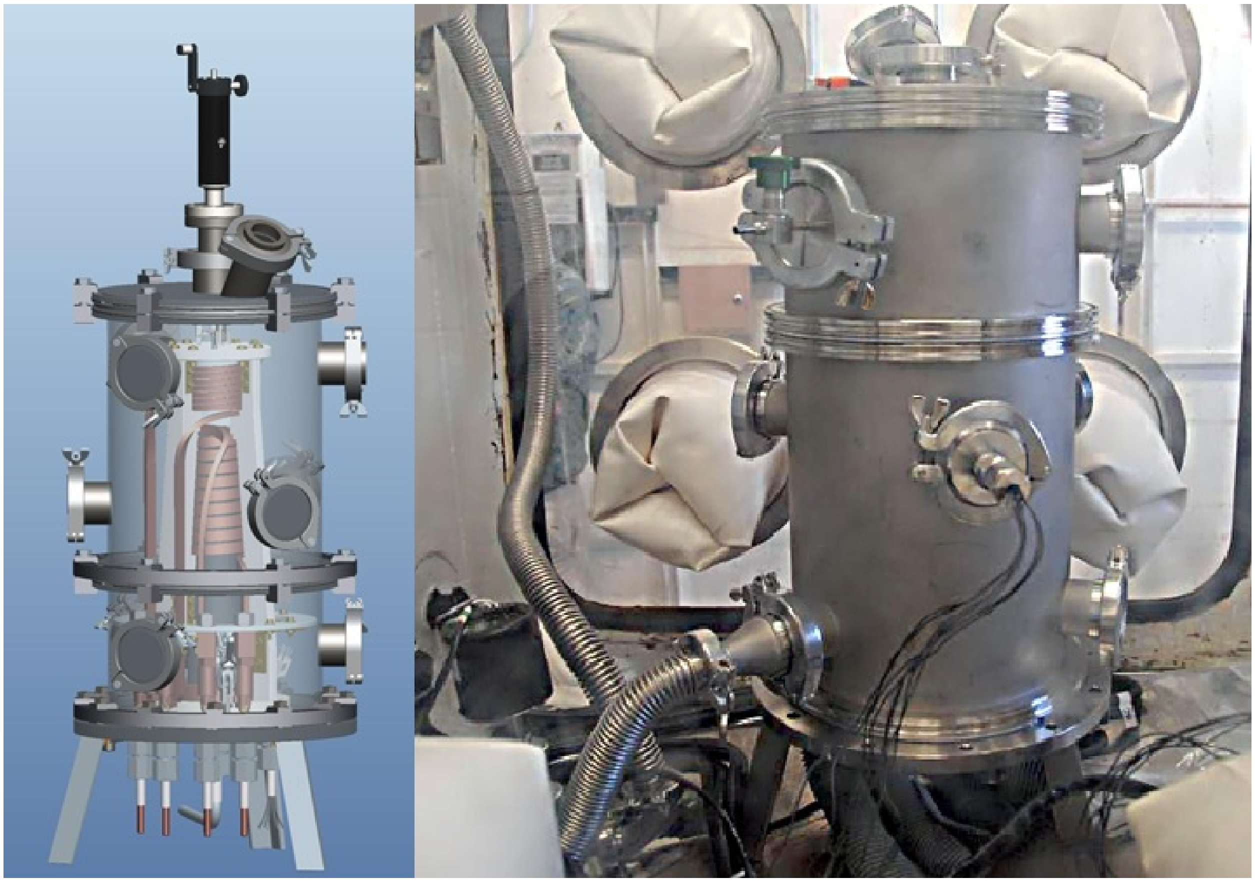

Figure 1. Left) Solid model of the GACS furnace Right) GACS furnace installed in the Casting Laboratory glovebox

2. CASTING TRIALS

Casting trials consisted of casting two separate U-10Zr alloys into the standard ZrO_2 slurry coated graphite mold and development of casting into integral FCCI barriers.

2.1 U-10Zr Casting

Previous U-10Zr castings were heated to approximately 1450°-1500°C and held for 10-15 minutes before pouring into the molds. Because the thermal differential was seen in the melt and crucible the crucible temperature at the time of casting was increased to 1600°C, in order to provide a melt temperature of at least 1500°C, well above the liquidus of U-10Zr. Table 1 shows the masses of the charges loaded. Both zirconium and uranium were loaded as elemental feedstocks, the uranium was in the form of 0.5" diameter rods of various lengths up to approximately 1 in. long and the zirconium was obtained from excess EBR-II fuel fabrication feedstock and was in the form of approximately 0.125 in. thick machine turnings. Figure 2 shows the heating profile used in the casting cycle. The furnace was programmed to reach 1600°C at a ramp rate of 150°C/min, hold at this temperature for 10 minutes and then cool. Figure 2 shows some initial control issues up to approximately 500°, and an initial hold of less than 1600°C due to furnace control issues. During the hold time, the stopper rod is lifted 30 seconds before the end of the cycle. If a casting is successful, once the stopper is raised a temperature rise is seen in the mold thermocouples, however, as seen in Figure 2 no rise was seen.

After cooling the furnace was opened and taken apart. The stopper rod was withdrawn and most of the zirconium that was originally charged had adhered to the stopper rod, although the uranium ran into the mold. After the crucible was removed it was seen that the majority of the uranium did not flow into the mold cavities, but rather stayed at the mold opening. Some small amount of material did flow into the molds producing small rods or pellets. Figure 3 shows the resulting stopper rod with adhered zirconium and residual melt and the cast products. One of the small rods does indicate that material did flow to the bottom of one mold cavity, but the flow was not a laminar consistent flow. Because this type of flow was not developed it is possible that the small amount of material flowed to the bottom of the mold, filled the vent there, but may not have expelled all of the needed inert gas from the mold. This action created too great of a pressure for further material to flow into the mold. This scenario has been seen in the past and produced segmented pins or rods. Also, earlier modeling work has verified the importance of having proper venting in place to avoid these segmented pins. The reason for not more fully flowing into the other mold cavities is not well understood. A possible contributing factor may be the amount of zirconium on the stopper rod may have slowed the flow down into the mold giving the material time to cool before it flowed more fully into the cavities. This is at least partially supported by the lack of mold temperature rise usually seen in the top portion of the mold when a large amount of material pools in this area.

Table 1. Elemental charges used in casting runs.

	1/21//2015 U-10Zr	2/12/2015 U-10Zr	2/24/2015 U-10Zr Zr Sheath	2/25/2015 U-10Zr Zr Sheath	8/10/2014 U-6Zr Zr sheath
Depleted Uranium	163.369	154.798	157.117	155.12	157.996
Zirconium	18.456	17.281	17.455	17.078	10.087
*Ending Process loss	0.099	0.126	0.116	0.43	0.384
*- Process loss is defined as material that is not recovered regardless of form					

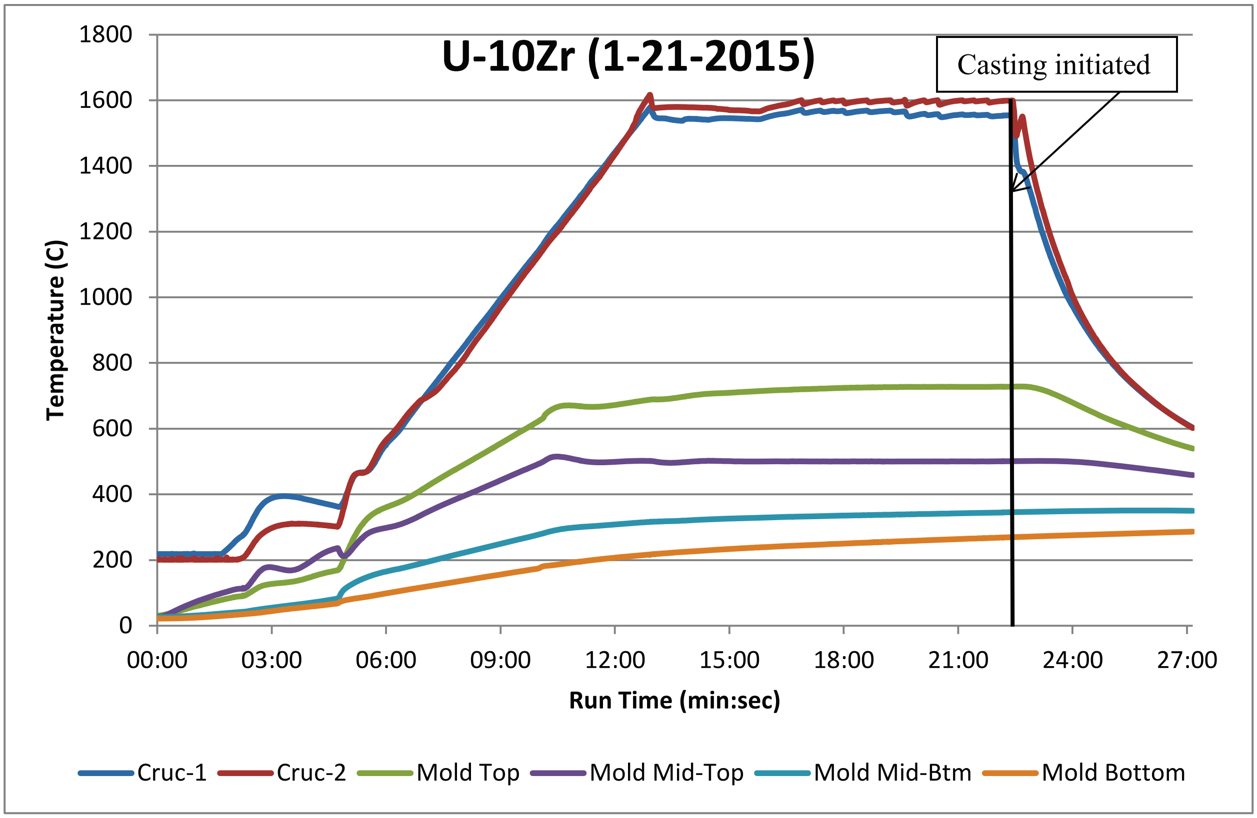

Figure 3. Heating profile for the 1-21-2015 U-10Zr casting run, notice no mold temperature rise is seen at the time of actual casting.

Figure 2. Resulting product from the 1-21-2015 casting attempt. Arrows indicate typical examples of undissolved Zr feedstock.

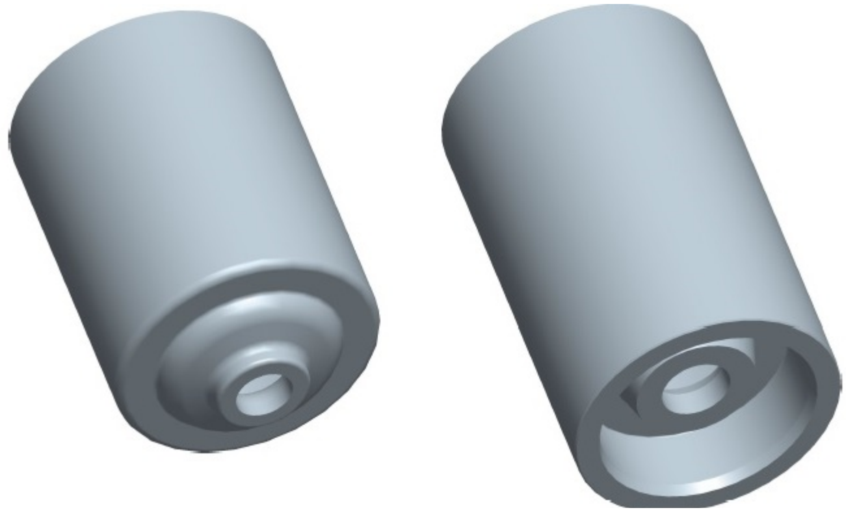

Figure 4. Crucible comparison between the a) earlier "non-locking" crucible and the b) locking or skirted version.

As mentioned in the previous year's report, a successful U-10Zr casting occurred early after the GACS was installed in the glovebox but many samples have had these dissolution or thermal gradient issues since then. This initial run was examined in greater detail with the only major change being the exterior shape of the lower portion of the crucible. The initial crucible used had a radius corner which interfaced with the top of the crucible. During mock up testing of the furnace however, the crucible exterior was modified with a "skirt" which locks the crucible onto the mold. This modification was made in order to facilitate easier assembly of the furnace. Sketches of both crucibles are shown in Figure 4. Because the skirted crucible was not available during the early glovebox operations of the GACS earlier non-skirted crucibles were used for the original casting experiments, and when the skirted crucibles became available they were incorporated. Based on this observation another U-10Zr casting run was done on 2/12/2015 which employed a non-skirted crucible. In order for a direct comparison to be made against the 1-21-2015 casting the casting process was configured as follows: the charge was similar, shown in Table 1, with the exception of some minor control issues at <400°C and a more consistent hold temperature heating profile was the same e.g. ramping to 1600°C at approximately 150°/min, holding for 10 minutes, and lifting the stopper rod at 9:30 of the hold time.

The resulting cast product was much better for this run. During the casting operation a very definite mold temperature rise was seen along the entire length of the mold at the time of casting indicating material flowed into the mold cavities. When the furnace was disassembled it could be immediately seen that the crucible had fully drained with only a small amount of melt and dross remaining. The dross was easily removed from the crucible and examined. There were no indications of undissolved zirconium remaining in the crucible, in the dross, or adhered to the stopper rod. The resulting cast products and crucible residue is shown in Figure 5. As can be seen, all pins flowed to the bottom of the mold cavities as evidenced by the flared ends on the right side of Figure 5. A void is seen approximately 5 in. from the bottom of one pin and the upper 1-2 in. have several flaws resulting in two of the pins breaking off of the mold heel approximately 9" from the bottom. The longest pin shown in Figure 5 is approximately 10 in., which is the full length of the mold. Diameters of the pins range from 3.93 mm (0.155 in.)-3.63 mm (0.143 in.) along the length. The flaws at the top of the pins do indicate that the vent may be a little smaller than is needed, thus some gas was not pushed out the mold but rather allowed to bubble up through the material in the mold cavity. Also, the large heel on top of the mold shows the charge was a little larger than needed.

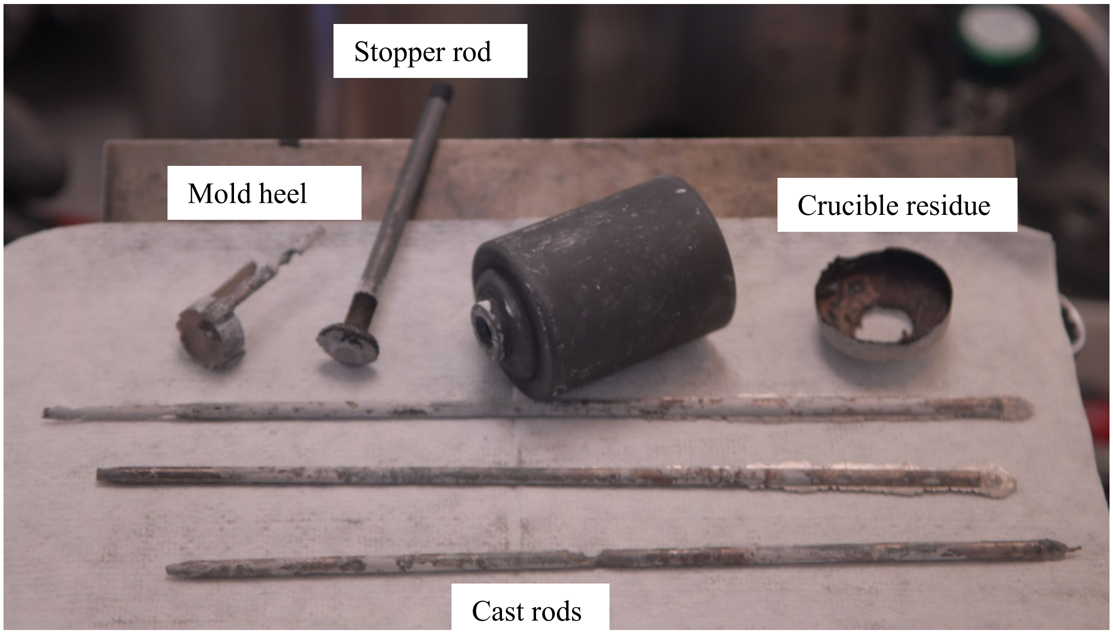

Figure 5. Crucible and resulting casting products from the 2-12-2015 casting run.

2.1.1 Crucible Thermal Analysis

The results above, along with the GACS configuration data, were transmitted to the Drs. Cetin Unal and Neil Carlson at Los Alamos National Laboratory for thermal analysis and simulation. Because both runs used the same temperature profiles the simulations looked at the steady state thermal conditions of the mold and crucible during the 1600°C hold period when both the mold and crucible are at constant temperature. The analysis compared the furnace thermal profile using the two different crucibles. Boundary conditions were set using actual measured temperatures for the mold and crucible. The thermal conductivity of both molds was assumed to be the same, 300 W/m^2. Initial simulations looked at only the melt temperature taking into account the mold temperatures provided through the run logs. The results of these simulations showed a 50-60°C difference in melt temperatures, with the skirted crucible being cooler, despite the crucible wall and mold mid top location being at the same temperature for both runs. These results are shown graphically in Figure 6. Simulations were continued to determine the temperature of the crucible/mold interface, which is not monitored during the run. These results are shown graphically in Figure 7. In these simulations it was shown that the top of the mold was approximately 47°C hotter for the skirted crucible. Based on these simulations an increase in heat flux from the skirted crucible to the mold of 18% was calculated compared to the heat flux from the non-skirted crucible to the mold. The increased heat flux leads to a melt that is generally cooler while the mold top is generally hotter.

In support of the analytical results obtained by Dr. Unal and Carlson the mold temperatures and power levels of the two runs were examined in more detail. The required power levels to maintain the casting temperature of the crucible and mold were compared. The crucible took approximately 10% less power to maintain the casting temperature of 1600°C in the case of the non-skirted crucible. The case of the mold is opposite with the non-skirted mold needing more power to maintain the casting temperature. This difference in the power requirements is evidenced by the differences in the mold temperatures along the length of the mold. Figure 8 compares mold temperatures of the two runs. As seen in Figure 8, on the mold top, this is approximately 8 in. from the mold base, the skirted crucible has a higher temperature as predicted by the thermal analysis. The mold mid-top thermocouple is located approximately 6 in. from the mold base. In this case both temperatures are the same. However, further down the mold, both the mid bottom thermocouple, 4 in. above the base, and the mold bottom thermocouple, 2 in. above the base, the skirted crucible case is cooler. These results all support the analysis showing the increased heat flux from the crucible to the mold in the case of the skirted crucible. Because of the increased heat flux going to the mold to maintain the casting temperature additional power is needed in the crucible in the skirted design. When the mold is examined the increased heat flux then will cause the mold top to heat up more. The mid-top thermocouples having the same temperature regardless of the crucible design is expected because this thermocouple has been selected to control the mold temperature. Therefore, the control system will control this temperature regardless of the power levels needed, independent of the other mold locations. Because of the increased heat flux with the skirted crucible, less mold power is required to maintain the casting temperature, as was seen in the run. This reduced power will result in the bottom two thermocouples being cooler when compared to the non-skirted crucible case because the lower power required to maintain temperature at the control thermocouple location reduces the temperatures lower in the mold. Based on the analytical results performed by LANL researchers, the experimental results, and observations made during the casting operations the crucible configuration definitely affects the thermal differential seen in the earlier castings. In order to avoid this issue future crucibles and crucible design modifications will ensure minimal contact between the mold and crucible.

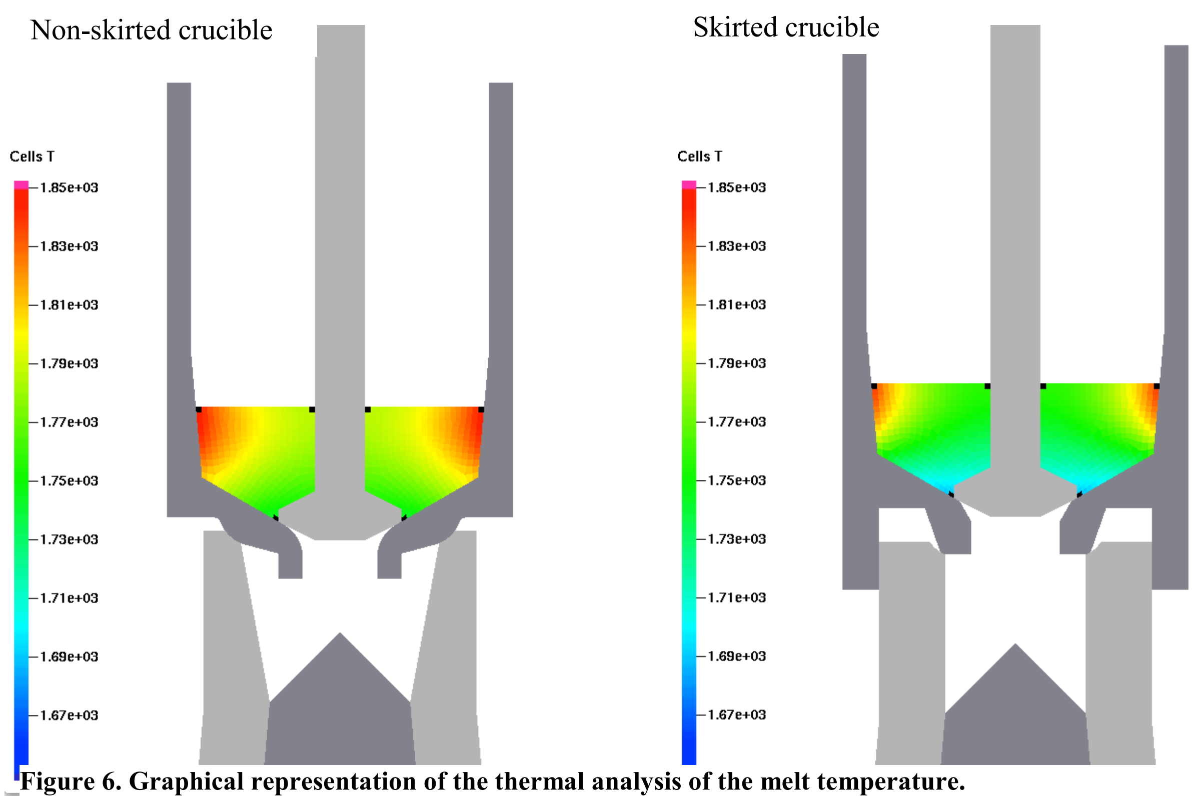

Figure 6. Graphical representation of the thermal analysis of the melt temperature.

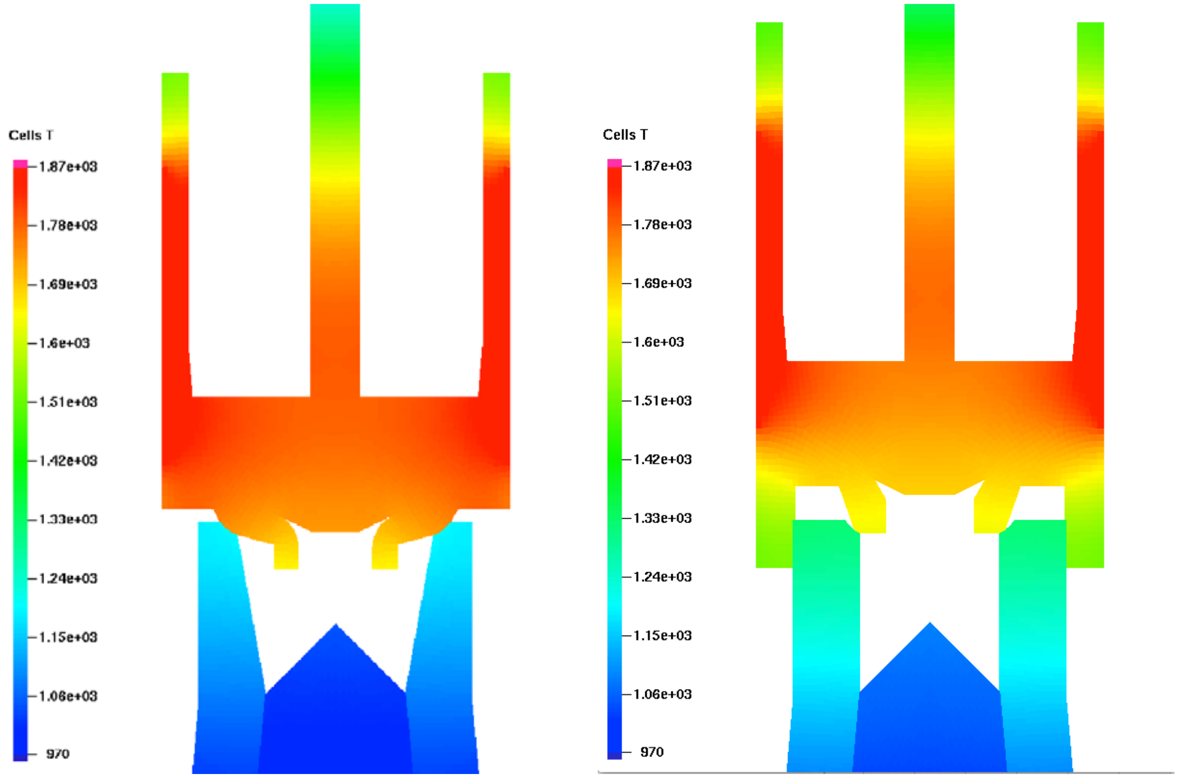

Figure 7. Graphical analysis showing the mold temperature on the skirted crcuible is approximately 47°C higher than the non-skirted crucible.

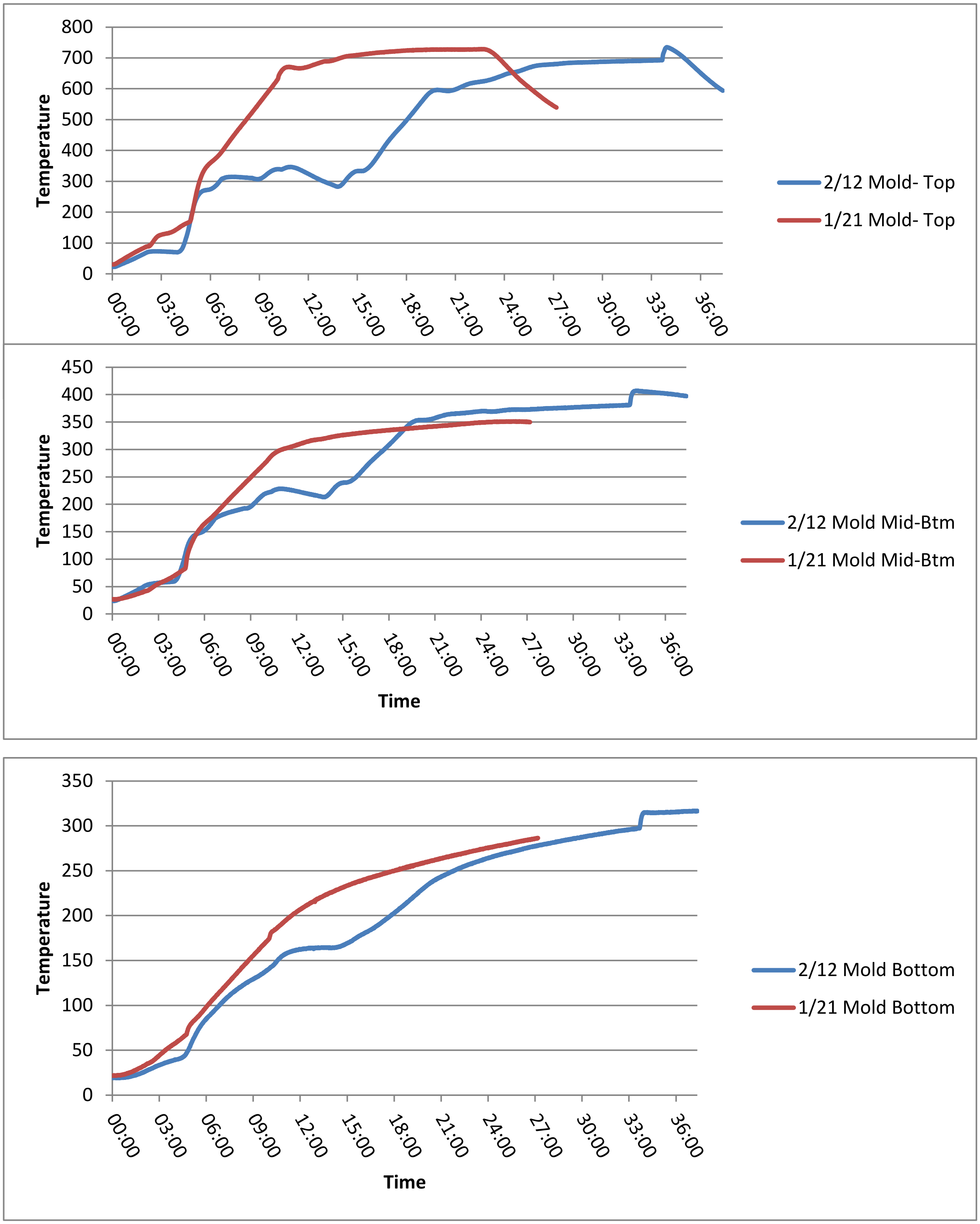

Figure 8. Comparison of mold temperatures throughout the casting runs. Note: 2/12 was done with a non-skirted crucible and 1/21 was performed using a skirted crucible.

3. CONCLUSION

In conclusion, casting optimization work is progressing. There were several months of inactivity due to glovebox maintenance, facility radiological control issues, and radiological source reduction and decontamination activities. Despite these periods of inactivity optimization activities progressed in the area of U-10Zr casting, crucible design activities, incorporation of an integral FCCI barrier, and permanent crucible coating.

Through experimental and analytical simulation work it was determined that because of the large contact area between the mold and crucible a thermal gradient was present in the melt. This gradient was large enough it impeded the dissolution of the zirconium. After the crucible design was modified to minimize the contact area casting quality improved. Work remains to be done to determine the optimal amount of superheat, application of vacuum pressure at the end of casting, or improved venting. These experiments will be done throughout next year, along with other experiments to determine the fluid properties of the melt and dependence on composition. Dimensional stability also will be investigated. Current fuel diameters are smaller than would be acceptable for a fuels test and is fairly variable over the length of a fuel pin. Mold cavities will be modified to enlarge the diameter as well as be more carefully characterized to ensure the cavities are consistent the length of the cavity.

Related to casting optimization is incorporation of the integral FCCI barrier. This year's work has shown that the barrier is feasible, although the minimal thickness has not been determined. As part of this optimization the amount of bonding seen between the alloy and sheath will be investigated. Because the protection concept depends on the sheath accommodating the fuel swelling a loosely bonded fuel would be preferable to a tightly bonded sheath/fuel interface. This interface strength may be affected by surface condition of the sheath i.e. oxidized, polished, etc., casting temperature, and composition. These areas will need to be further examined and optimized through additional experiments and a large amount of characterization. As with bare casting optimization some dimensional optimization is also necessary. The current batch of pins showed an excessive amount of diametral variation and were undersized. The dimensions are of course influenced by the mold dimensions but also the sheath dimensions. Sheath fabrication methods may also be investigated to ensure the correctly sized sheaths are used in the casting operations.

In the area of re-usable crucible coating development the initial "base case" has been performed but is awaiting further characterization. Visually the coatings appear to be intact with only minor discoloration. Further microstructural characterization work will be necessary to verify the coatings are still intact and are not experiencing excessive degradation. Further tests will be conducted which will incorporate rare earth elements to simulate a recycled fuel product. Many of the rare earths are chemically more aggressive than the actinides and zirconium, and because the current Y_2O_3 coating is itself a rare earth it may be susceptible to continual break down. Also, the base case included only one exposure cycle; a re-usable coating will be subjected to several cycles and therefore must also be tested for several cycles.