

**Project Title:****Design and Analysis of a Process for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides****April 12, 2002**

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**Abstract**

UNLV has developed and will continue to develop process models for the analysis of melt casting processes. This work will continue to be performed under the guidance of Argonne National Laboratory (ANL) engineers to ensure that their knowledge and experience benefits the project. The research to be conducted during the second year will center on performing detailed analyses on a conceptual design of an inductively heated skull-crucible casting furnace. Processing conditions will be analyzed, basic models utilized, and detailed heat and mass transfer models will be developed to analyze the most promising processes. The goal of this second year is to complete the preliminary design analyses for the proposed skull-crucible casting furnace, which will be developed and tested in subsequent years.

## Accomplishments – Year 1

Reviewed and selected a proposed casting furnace design that could potentially minimize americium vaporization. Models of the important physical processes were also developed: mold filling and heat transfer, mass transport, and induction heating.

## Proposed Work – Year 2

UNLV is uniquely qualified to perform this work because of resident expertise in the analysis of engineering systems. Research efforts will focus on the development of complex heat transfer, mass transfer, and inductive heating models.

### 1. Progress in Year 1

#### 1.1 Selection of Melt Casting Process

A review of existing literature was undertaken to select potential casting technologies. The casting process variables are shown in Figure 1. These technologies were then evaluated to select one melt casting process that was felt to have the greatest chance to decrease americium transport. Several candidate designs were proposed and evaluated in collaboration with Argonne National Laboratory staff. An inductively heated skull-crucible design was ultimately selected and is shown in Figure 2. This design offers realistic processing times, potential confinement options to minimize americium transport, reasonable purity levels, and there is experience within the DOE complex and ANL-West to facilitate the testing of such an apparatus.

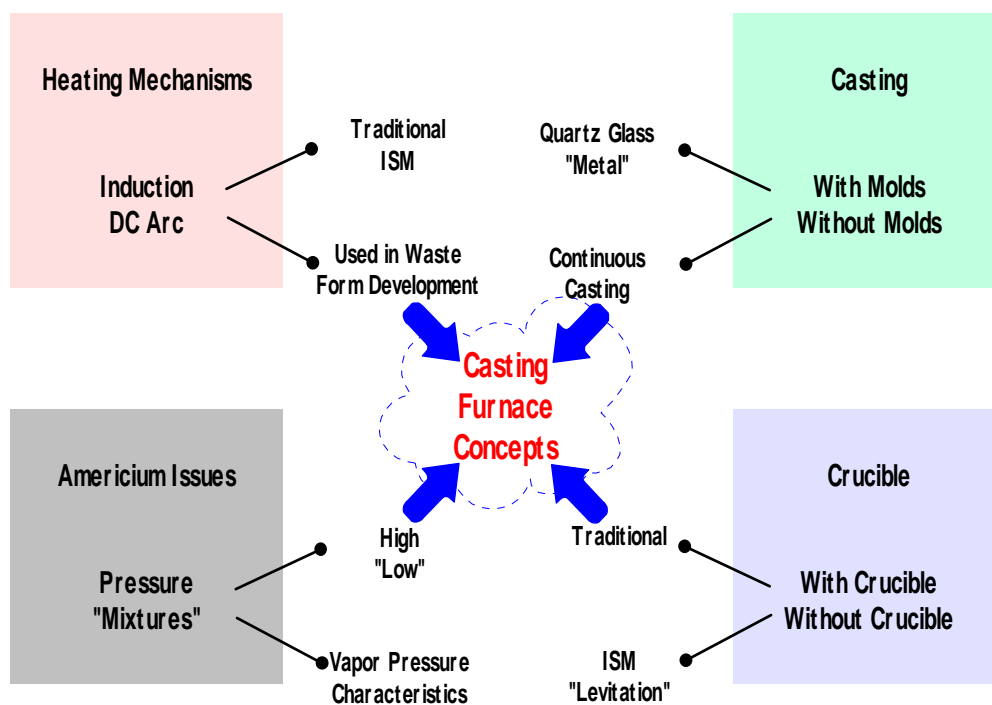


Figure 1. Casting furnace process variables

## 1.2 Parametric Model for Fuel Rod Casting

In producing an acceptable fuel rod, there are two issues that needed to be addressed: the retaining of americium and the casting of a long slender rod. A model has been developed and analyses are currently being completed to assess different mold designs, different operating conditions (preheating and superheat), and different aspect ratios.

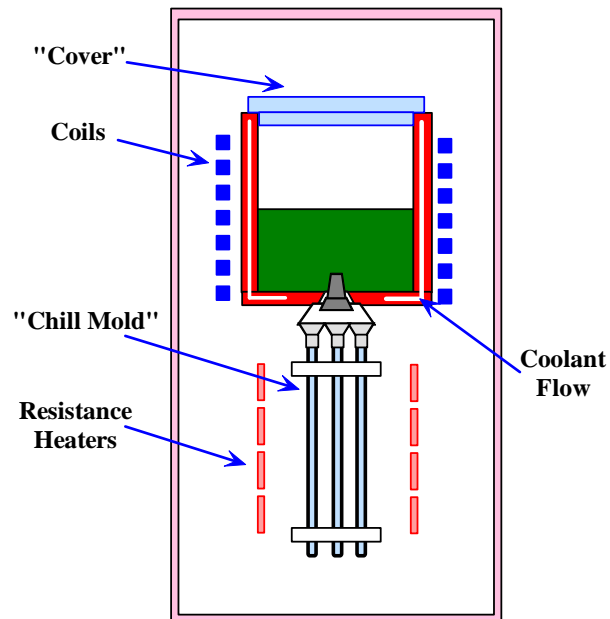


Figure 2. Inductively heated skull-crucible melting design (ISM)

Typical results for the modeling effort are shown below. Figure 3 shows the flow field moving down through the mold. The outer mold is considered to have the properties of copper. Figure 4 shows preliminary estimates of the pressure at the inlet of the mold region. This information will be useful to assess which techniques are suitable for casting long slender fuel rods.

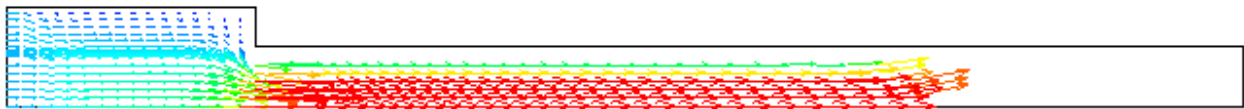


Figure 3. Typical flow entering the mold region

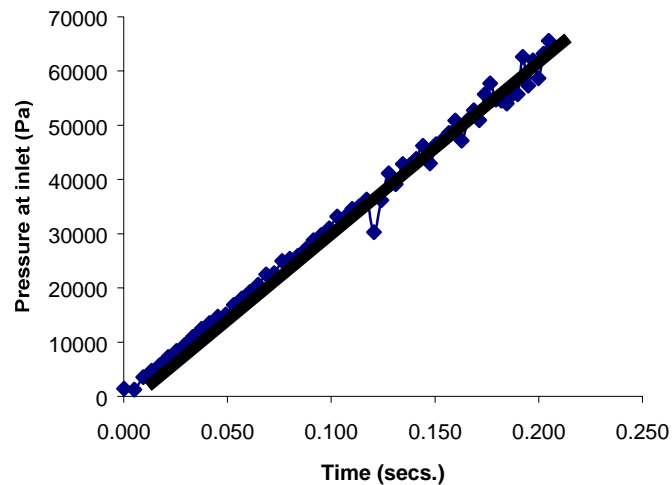


Figure 4. Inlet pressure for flow into the mold region

At present, the finite element package FIDAP is used for all of the analyses shown here. The complex nature of the modeling effort has uncovered some subtle model deficiencies within FIDAP. Developers, support specialists, and UNLV personnel are working to resolve these deficiencies. Once these deficiencies have been addressed to our satisfaction, parametric studies will be carried out to assess a number of process parameters.

### 1.3 Development of a Mass Transfer Model

A useful reference was obtained through discussions with ANL-West staff and our preliminary literature search. The reference outlines a reasonable approach to estimating the mass transfer of a volatile metal constituent in an alloy system. The approach:

- Documents theoretical relationships for the estimation of activity coefficients,
- Outlines the transport mechanisms for transport and vaporization of the volatile metal,
- Analyzes the impact of chamber pressure on the transport rates, and
- Predicts the proper trends.

The modeling approach has been selected and efforts for the remainder of the first year's activities will center on developing and implementing this model for americium in a plutonium/zirconium system.

In an i-j binary system, the relationship between the activity coefficient  $\gamma_i$  of the component i and its partial mole excess free energy  $\overline{G}_i^E$  can be written as

$$\overline{G}_i^E = RT \ln \gamma_i$$

in which

$$\bar{G}_i^E = G_{ij}^E + (1 - x_i) \frac{\partial G_{ij}^E}{\partial x_i}$$

where  $G_{ij}^E$  is the system's excess free energy and is given by

$$G_{ij}^E = \Delta H_{ij} - TS_{ij}^E$$

An approximate relationship between the heat formation of the binary system  $\Delta H_{ij}$  and its excess entropy  $S_{ij}^E$  will be calculated in the second year tasks as follows:

$$S_{ij}^E = 0.1 \Delta H_{ij} \left( \frac{T_{mi} + T_{mj}}{T_{mi} T_{mj}} \right)$$

where  $T_{mi}$  and  $T_{mj}$  are the melt points of components i and j, respectively. If we define

$$\beta_{ij} = 1 - 0.1T \left( \frac{T_{mi} + T_{mj}}{T_{mi} T_{mj}} \right)$$

then

$$G_{ij}^E = \beta_{ij} \Delta H_{ij}$$

The activities of Am in molten U-Am and Pu-Am will be calculated in the second year. The model will include transport of the species to the surface of the melt, which are mainly the diffusion process of evaporating elements, vaporization at the liquid-gas interface, transport of the vapor in the gaseous phase, and the condensation of the species on surrounding surfaces. The controlling steps of the mass transfer process will also be studied in the second year tasks. Previous research efforts were focused on the transport to the melt surface and the evaporation at that interface. The additional two stages of transport are also important in the present work. The transport of the vapor through the gas phase and eventual deposition on surrounding surfaces is important. Knowing where the americium will deposit or where it will be transported is a major concern.

Efforts through the remainder of the first year will finalize this model and demonstrate its utility.

## 1.4 Development of an Induction Heating Model

The melting and flow of materials within the crucible depends directly on the amount of induction heating and stirring due to the magnetic fields. In order to account for these effects a model is being developed to predict the amount of energy deposited in conducting materials. Once this model has been developed, further enhancements can be made to include the “stirring” effect in the fluid.

Starting with Maxwell’s Equations and making realistic assumptions for a cylindrical coil leads to the following set of equations to be solved in the different regions:

$$\left. \begin{aligned} \nabla \cdot \left( \frac{1}{r} \nabla C \right) &= -\mu J_o \\ \nabla \cdot \left( \frac{1}{r} \nabla S \right) &= 0 \end{aligned} \right\} \text{Coil}$$

$$\left. \begin{aligned} \nabla \cdot \left( \frac{1}{r} \nabla C \right) &= \frac{\mu \sigma \omega}{r} S \\ \nabla \cdot \left( \frac{1}{r} \nabla S \right) &= -\frac{\mu \sigma \omega}{r} C \end{aligned} \right\} \text{Conductor}$$

$$\left. \begin{aligned} \nabla \cdot \left( \frac{1}{r} \nabla C \right) &= 0 \\ \nabla \cdot \left( \frac{1}{r} \nabla S \right) &= 0 \end{aligned} \right\} \text{Elsewhere}$$

where

- C, S = real and complex components of function substituted into governing equations to simplify solution process
- r = radial coordinate
- $J_o$  = current density
- $\mu$  = permeability
- $\omega$  = frequency
- $\sigma$  = electrical conductivity

Using the appropriate relationships and integrating gives the heat deposition as a function of position.

$$Q(r, z) = \frac{\sigma \omega^2}{2r^2} [S^2 + C^2]$$

The quantities  $S$  and  $C$  can be used to back calculate other field quantities as well. These equations are currently being solved for a test problem previously analyzed and published in the literature. This will allow the validation of the implementation of these equations. A first attempt is being made to solve these equations in the CFD code FIDAP, which would eventually allow for a solution to the heat transfer, mass transfer, and fluid mechanics aspect of the crucible

design. Complete details of the derivation and validation will be part of the first year's annual report.

## **2. Research Objectives – Year 2**

There are four research objectives for the second year of this project. These objectives are to:

1. Continue to refine and assess americium transport issues within the furnace. This will include collaboration with other national laboratory researchers to better define the physical and chemical properties of americium.
2. Develop an integrated model for a skull-crucible melt casting furnace.
3. Perform preliminary design analyses to support the development of lab scale furnace in future years.
4. Review and select potential surrogate materials for future testing in skull crucible furnace and model evaluation.
5. Design and begin to perform experiments necessary for code validation.

Work needs to continue related to the properties of americium. Characterizing its physical properties is still a key to effectively modeling the transport of americium in the casting furnace. A model based on fugacity will be completed in the first year. Developing and testing the limitations of this model will help us determine what physical quantities still need to be obtained (numerically or experimentally) in order to improve our models.

The most difficult objective is incorporating all physical phenomena into a numerical model. The physics of importance include induction heating, mass transfer, heat transfer, and fluid mechanics. Integrating as many of these physical phenomena as possible will aid in the assessment of which process parameters could be modified to decrease americium transport.

Once a detailed model has been developed, the preliminary design of an inductively heated skull-crucible will be undertaken. This work will be carried out with the guidance and support of ANL-West staff. The ANL-West staff has experience with skull crucible furnaces and they have existing hardware that could potentially be modified for testing with surrogate materials. Using existing hardware could potentially decrease costs and shorten development times.

The fourth objective is to develop a list of potential surrogate materials that could be tested in a glovebox facility to aid in model validation and to assess the proposed system design. The surrogate materials would have to have qualitatively similar properties from a mass transfer standpoint.

The last objective is to design and begin to perform code validation experiments using surrogate materials. Experiments are required in the areas of a) solidification of a long rod in a chill mold, b) behavior of volatile species in a skull melter, and 3) coupling of the induction field to the charge in a skull melter.

### 3. Technical Impact

The AAA program requires a non-fertile actinide form to serve as the “fuel” for the transmuter blanket. The currently proposed candidates for this fuel form still include a metallic alloy fuel, a cermet fuel, and a nitride fuel. Each of these candidates has been proposed based on known performance of the fertile fuel (i.e., uranium) analogue.

Of primary concern to the AAA program is the requirement for fabrication of the selected fuel form in a remote, hot cell environment. The second year of funding for this activity will be used to develop a preliminary furnace design that can be built and tested with surrogate materials in future years. The key to assessing the viability of metal fuels is in the ability to demonstrate fabrication in the presence of a volatile constituent.

The groundwork laid in year one of this project developed a set of modeling tools to assist in the design of a realistic fabrication technique. The primary technical hurdle to overcome in the fabrication of a metallic alloy fuel is that of efficiently including the highly volatile actinide elements (i.e., americium).

As stated in last year’s proposal, previous attempts at incorporating americium into metallic alloy fuels using standard injection casting processes have resulted in the volatilization and loss of up to one-half of the initial americium charge introduced into the melt. This magnitude of loss due to volatilization is unacceptable, and would almost certainly exclude metallic alloy transmutation fuels from consideration if an engineering solution cannot be developed in the near term. Our goal is to assist in making this assessment, plus training students in the use of complex physics models related to the nuclear industry.

### 4. Research Approach

The proposed research has been broken down into six specific tasks. These tasks are outlined below.

1. *Collaborate with other National Laboratory Researchers:* Efforts will be made to collaborate with researchers at national laboratories to obtain physical property data for americium. Research efforts are under way by other researchers to numerically predict the physical properties of a number of important materials.
2. *Parametric Modeling of Volatile Actinide Transport:* Work will continue in this area during the second year. These sensitivity studies will aid us in determining what physical parameters are needed in order to more accurately model the mass transport issues.
3. *Integration of Multiple Physics into Numerical Model:* Traditionally, inductively heated furnaces have been designed through the use of one-dimensional calculations, or “rules-of-thumb.” In order to more accurately assess the impact of process parameters on the transport of americium, more detailed models are needed. These detailed models, used in conjunction with the knowledge of experienced designers, will improve our understanding of furnace operations. Efforts will be made to incorporate heat transfer, mass transfer, fluid mechanics, and induction heating into one model.

4. *Preliminary Design of Furnace.* This effort will be carried out in conjunction with ANL-West staff. Key components of an existing skull-crucible system already exist at ANL-West including component information and the experience of ANL-West staff will be crucial in designing a system that meets the needs of the AAA program.
5. *Review and Selection of Surrogate Materials:* Potential materials will be identified that could be used to represent the volatile nature of americium. These materials will have to be safe for glovebox use and be able to be heated inductively.
6. *Reporting Requirements:* Monthly activity updates will be submitted to the UNLV AAA program director as required by DOE management. Quarterly progress reports will be completed and filed with the UNLV AAA program office in addition to being sent to ANL collaborators. Researchers will publish the results of this project at the appropriate technical conferences and journals. An annual report will be compiled and submitted for review as well.

Dr. Clarksean's work has been crucial in developing an accurate model that properly represents the physics of the problem. His efforts have centered on working with the software developers at FLUENT in resolving and developing proper modeling techniques for the VOF method used within FIDAP for the accurate modeling of the flow. Properly modeling the flow, has led to more accurate heat transfer models.

In addition, Drs. Clarksean and Chen have developed and implemented an induction-heating model within FIDAP. This is a critical component in the complete analysis of the proposed casting furnace. To complete this work it was necessary to derive the governing equations in their proper form, develop a procedure that allows solution of the equations in rectangular coordinates, but at the same time allows for the results to be implemented into a cylindrical coordinate model. Proceeding in this fashion allows one code to be used for the calculation of the induction heating, fluid flow, and heat transfer.

Drs. Chen and Clarksean have been working to develop a detailed model of the mass transfer from the melt. This model is the critical component of the complete modeling process. The transport of americium from the melt to the surrounding areas is the most important aspect of the project. Developing this model in a fashion that allows the mass transfer to be included along with all of the other important physics is difficult.

The complete model requires the solution of several sets of coupled equations. The equations include:

- Navier-Stokes equations for the flow of an incompressible fluid (two momentum and one continuity equation). These equations will eventually be coupled to the energy and induction heating equations (body force term).
- Mass transfer equation for the transport of americium.
- Heat transfer equation for heating and cooling of the melt material.
- A set of coupled second order partial differential equations for the real and imaginary parts of the induction heating equations (highly nonlinear and strongly coupled – stiff).
- Plus the setting of a number of important physical boundary conditions.

The solution of these equations is tough numerically and very computationally intensive. Drs. Clarksean and Chen both have the knowledge and experience needed to attempt the solution of this complex problem. Limited research has been done on these complex materials processing problems. Drs. Chen and Clarksean will not only supervise and help graduate and undergraduate students to work on this research, but also serve themselves as research engineers on this project to develop the furnace design for metallic fuel pins.

## **5. Capabilities at UNLV and DOE Labs**

Dr. Yitung Chen is a Research Associate Professor of the Department of Mechanical Engineering and Interim Director of the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada, Las Vegas, and serves as Principal Investigator. He received his B.S. degree in Chemical Engineering in 1983, and his M.S. and Ph.D. degrees in Mechanical Engineering in 1988 and 1991, respectively, from the University of Utah. He also has a minor degree in Nuclear Engineering. He was a consultant for several engineering companies from 1991 to 1993. Dr. Chen is an expert in experimental and computational aspects of momentum, heat, and mass transfer. His research interests include chemical kinetics modeling, high level radioactive waste repository design, atmospheric sciences, magneto-hydrodynamics modeling, ground water transport, energy conservation, and biomedical engineering. He also has a strong background in organic chemistry, biochemistry, polymer chemistry, and physical chemistry. His research experience includes being PI and co-PI on projects involving the study of flow and heat transfer and species transport in unsaturated porous media funded by DOE, the burning of rocket motors under the Joint Demilitarization Technology (JDT) program funded by DOD, and atmospheric modeling funded by the NOAA Cooperative Institute for Atmospheric Sciences and Terrestrial Applications. He is also co-PI on an EPA project dealing with environmental monitoring for public access and a groundwater modeling project funded by DOE.

Dr. Darrell Pepper is Interim Dean of the College of Engineering and Director of the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada, Las Vegas. Dr. Pepper serves as Co-Principal Investigator. He has been actively involved in the generation, development, and use of hybrid, multi-dimensional algorithms for environmental transport and CFD applications for many years, and has developed atmospheric models for the NRC, NOAA, and DOE (OHER; NVOO). His previous work experience at the Savannah River Site (E. I. Du Pont de Nemours), the Marquardt Company, and Advanced Projects Research, Inc., have resulted in numerous publications and presentations. Dr. Pepper organized and directed the first modeling workshop for the DOE-OHER on mesoscale atmospheric transport modeling. He served on a NRC project to assess consequences of natural phenomena on various reactor sites and fuel fabrication facilities located within the U.S., and has developed 3-D dispersion models for the NRC and DOD. Dr. Pepper is the co-author of three textbooks on finite element methods, co-editor of two books on environmental modeling, and directs the AIAA Home Study Courses and ASME short courses on finite elements. He is a Fellow of ASME and Associate Fellow of AIAA.

Dr. Randy Clarksean's experience in process modeling and the casting furnace process currently employed at ANL provides a solid base from which to devise and evaluate new casting furnace concepts under the unique conditions that exist with volatile actinides. He is familiar with

current casting furnace operations at ANL and has previously developed first order models of the system. Dr. Clarksean completed his Ph.D. at the University of Utah in 1990, with an emphasis on computational methods in the thermal and fluid sciences. In 1990, he started work for Argonne National Laboratory at their Idaho facilities. He worked on a number of different process and safety projects while with ANL. Since 1995, Dr. Clarksean has worked on a number of projects independently. These projects have involved materials processing, spent nuclear fuel storage, electronics cooling, phase change, and other general heat and mass transfer processing. Funding for these projects have come from the DOE, DOD, private industry, international research organizations, and the State of California. He is an expert in the analysis of engineering systems and has numerous publications in heat transfer and fluid mechanics.

ANL West has extensive experience in injection casting of uranium and plutonium alloys, having fabricated literally hundreds of thousands of metallic alloy fuel pins for irradiation in EBR-II using this process. Currently, two injection casting furnaces are in operation at ANL West, and previous experimentation attempting to incorporate americium into U-Pu alloys has been conducted. This past experience has led investigators at ANL West to hypothesize over the period of almost 10 years as to what process modifications or furnace design changes might improve the chances for successful melt casting of actinide alloys. The cooperation between the laboratories will provide a good working relationship to develop a solid approach to developing the next generation casting furnace for the AAA Program.

Extensive computing facilities exist for the modeling efforts at UNLV. Computing facilities range from workstations to super computers. These facilities will be available for the analysis and design of the proposed concepts. A wide range of computational tools exists on these systems.

## **6. Project Timeline with Milestones and Deliverables**

The proposed schedule for all tasks and significant meetings is shown on the following page. Three meetings are planned for the year.

Tasks:	Qtr 1			Qtr 2			Qtr 3			Qtr 4		
	June	July	Aug	Sept	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May
1. Collaborate with National Laboratories (americium properties)		▼	▨	▨	▨	▨						
2. Parametric Modeling of Volatile Actinide Transport	▼	▨	▨	▨	▨	▨						
3. Integration of Multiple Physics Into Model	▼	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨
4. Preliminary Design of Furnace and Experiments for Model Validation							▼	▨	▨	▨	▨	▨
5. Review and Selection of Surrogate Materials							▼	▨	▨	▨	▨	▨
7. Reporting Requirements				●			●			●		●
Travel to ANL for Meetings							◇			◇		◇
Notes:												
	1											
	2											
	3											
Trips 1 and 2	Visit ANL-West to discuss preliminary design of casting furnace (Task 4).											
Trip 3	Visit ANL-West to discuss modeling and research efforts prior to submittal of final report (feedback on work)											

**Figure 1 - Proposed Timeline for Research Tasks.**

Work is assumed to commence on June 1, 2002. Additional travel may be necessary for interactions relating to other specific tasks.