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Design and Analysis for Melt Casting Metallic Fuel Pins Incorporating Volatile Actinides: Quarterly Progress Report 5/16/02- 8/15/02

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UNLV-AAA University Participation Program

Principle Investigator: Yitung Chen
Co-Principle Investigators: Randy Clarksean and Darrell Pepper

Purpose and Problem Statement

An important aspect of the Advanced Accelerator Applications (AAA) program is the development of a casting process by which volatile actinide element (i.e., americium) can be incorporated into metallic alloy fuel pins. The traditional metal fuel casting process uses an inductively heated crucible. The process involves evacuation of the furnace. The evacuation of the furnace also evacuates quartz rods used as fuel pin molds. Once evacuated the open ends of the molds are lowered into the melt; the casting furnace is then rapidly pressurized, forcing the molten metal up into the evacuated molds where solidification occurs.

This process works well for the fabrication of metal fuel pins traditionally composed of alloys of uranium and plutonium, but does not work well when highly volatile actinides are included in the melt. The problem occurs both during the extended time period required to superheat the alloy melt as well as when the chamber must be evacuated. The low vapor-pressure actinides, particularly americium, are susceptible to rapid vaporization and transport throughout the casting furnace, resulting in only a fraction of the charge being incorporated into the fuel pins as desired. This is undesirable both from a materials accountability standpoint as well as from the failure to achieve the objective of including these actinides in the fuel for transmutation.

Candidate design concepts are being evaluated for their potential to successfully cast alloys containing volatile actinides. The selection of design concepts has been conducted in close cooperation with ANL staff. The research centers on the development of advanced numerical models to assess conditions that significantly impact the transport of volatile actinides during the melt casting process. The work will include the collection and documentation of volatile actinide properties, development of several conceptual designs for melt casting furnaces, modeling and analysis of these concepts, development of sophisticated numerical models to assess furnace operations, and analysis of these operations to determine which furnace concept has the greatest potential of success. Research efforts will focus on the development of complex heat transfer, mass transfer, and inductive heating models.

Personnel

Principle Investigator:

- Dr. Yitung Chen (Mechanical Engineering)

Co-Principle Investigators:

- Dr. Randy Clarksean (Mechanical Engineering)
- Dr. Darrell Pepper (Mechanical Engineering)

Graduate Student:

- Mr. Xialong (Frank) Wu, M.S. Graduate Student, (Mechanical Engineering)

National Laboratory Collaborators:

- Dr. Mitch Meyer, Leader of Fabrication Development Group, ANL-West
- Dr. Steve Hayes, Manager of Fuels & Reactor Materials Section, Nuclear Technology Division, ANL-West

Management Progress

Budget Issues:

- The graduate student and research faculty contracts were not prepared until the end of May because the research budget was not allocated before the end of April 2002.

Student Issues:

- Mr. Xiaolong Wu has successfully defended his master thesis on May 31, 2002.
- The graduate student, Mr. Taide Tan, of the Mechanical Engineering Department has been recruited to participate the research. He will apply his student visa in Guang Zhou, China in July.

Management Problems

No management problem issues at this time.

Technical Progress

The major accomplishment of this project in May 2002 was that our student Mr. Xiaolong Wu finished his master thesis defense on May 31. A few minor changes and revisions were made according to his thesis committee's suggestions.

Induction heating model and governing equations have also been developed. The modeling efforts have centered around the development of the governing equations, developing a method to incorporate then into FIDAP, setting up a test problem, and making preliminary calculations for the geometry of interest. We will present a paper

titled as "Development of a Model for Induction Heating," in the FIDAP User Group Meeting in Chicago.

The evaporation kinetics of americium from melt metals will be studied. We will continue to develop the mass transfer model and study kinetics and activities parameters of different actinides for the metallic fuel pins furnace design.

In order to test the impact of process parameters (temperature, pressure, alloying elements, etc.) on the casting process, a parametric study of the casting model was performed on different parameters to determine which process parameters are critical in manufacturing a suitable metallic fuel pin. For some specific properties, which are not available, it will parametrically vary these properties over the expected range to see how these properties might exhibit and determine how critical these properties are to the processing of the fuel.

Parameter study efforts centered around model development and the analysis of the impact of mold preheating on heat transfer into the mold. Inner radius is already set by the Argonne National Laboratory.

Normally, the way the heat flows across the metal and mold surfaces directly affects the evolution of solidification, and plays a notable role in determining the freezing conditions within the metal. When metal and mold surfaces are brought into contact an imperfect junction is formed. While uniform temperature gradients can exist in both metal and mold, the junction between the two surfaces creates a temperature drop, which is dependent upon the thermophysical properties of the contacting materials, the casting and mold geometry, the roughness of mold contacting surface, the presence of the gaseous and non-gaseous interstitial media, the melt superheat, contact pressure and initial temperature of the mold. Because the two surfaces in contact are not perfectly flat, when the interfacial contact pressure is reasonably high, most of the energy passed through a limited number of actual contact spots. The heat flow across the casting-mold interface can be characterized by a macroscopic average metal-mold interfacial heat transfer coefficient (h_i), given by

$$h_i = \frac{q}{A(T_{1C} - T_{1M})} \qquad (9)$$

The heat transfer coefficient shows a high value in the initial stage of solidification, the result of good surface conformity between the liquid core and the solidified shell. As the solidification progresses the mold expands due to the absorption of heat and the solid metal shrinks during cooling and as a result a gap develops because pressure becomes insufficient to maintain a conforming contact at the interface. Once the air gaps form, the heat transfer across the interface decreases rapidly and a relatively constant value of h_i is attained. The ways of heat transfer here are assumed to be due to both conduction through isolated metal-mold contacts and through gases present in the gap and radiation between the surfaces.

Previous research shows that the process of thermal energy stored in liquid metal to the mold at the metal-mold interface during solidification is determined by the heat transfer coefficient at the interface. The quality of the final product-fuel pin is directly affected by the interfacial heat transfer conditions, which is mostly from heat transfer coefficient.

Average fill velocity was also changed to see how do they affect the filling process. Tables through 1 and Table 2 show all the parameter values used under various simulation circumstance. Table 3 and 4 show all basic physical properties of mold and melt respectively.

Table 1 Parameters selected for copper simulation

	Mold length = 0.5 m		
	HT Coefficient = 2000,5000 W/m^2K		
Copper Mold	Mold Preheating Temperature (^oC)		
Inlet Velocity (m/s)	400	800	1200
0.1	×	×	×
1.0	×	×	×
2.0	×	×	×

Table 2 Parameters selected for quartz simulation

	Mold length = 0.5 m		
	HT Coefficient = 2000, 5000 W/m^2K		
Quartz Mold	Mold Preheating Temperature (^oC)		
Inlet Velocity (m/s)	400	800	1200
0.1	×	×	×
1.0	×	×	×
2.0	×	×	×

Table 3 Physical properties of mold materials

Physical Properties of Mold Materials	"Copper"	Quartz
Thermal Conductivity (W/m·^{o}C)	320	1.4
Specific Heat (J/Kg·^{o}C)	380	670
Density (Kg/ m^{3})	8933	2200

Table 4 Physical properties of melt

Melt Contents	Pu, Zr, Am, etc
Thermal Conductivity (W/m·K)	6
Specific Heat (J/Kg·^{o}C)	150
Density (Kg/ m^{3})	19800
Viscosity (N·s/ m^{2})	0.1

We have also studied the actinide of americium: Americium, element 95, was discovered in 1944-45 by Seaborg et al. at the Metallurgical Laboratory of the University of Chicago. The reaction used was:

$$^{239}Pu\,(n,\gamma)\,^{240}Pu\,(n,\gamma)\,^{241}Pu \xrightarrow[14\ yr]{\beta^-} {}^{241}Am$$

During all the isotopes of americium from ^{237}Am to ^{246}Am, only ^{243}Am (half-life = 7600 years) and ^{241}Am (half-life = 433 years) are sufficiently long lived to permit accurate property studies and are the most important and the most useful for chemical research.

Metal Preparation: Americium metal has been prepared by the following methods:

1. Reduction of AmF_3 with barium (or lithium) metal
2. Reduction of AmO_2 with lanthanum metal
3. Bomb reduction of AmF_4 with calcium metal
4. Thermal decomposition of Pt_5Am

Allotropy:Americium metal is silvery, ductile, non-magnetic and very malleable. There are three well-established forms of americium metal:

1. phase: double hexagonal close-packed structure
2. β phase: body-centered structure
3. γ phase: face-centered cubic structure

Thermodynamics of Americium Metal Transitions

Transition	T(K)	ΔH_t(KCal·mol^{-1})	ΔS(Cal·K^{-1}·mol^{-1})
α→β	923±50	0.185	0.2
β→γ	1349±5	1.400	1.04
γ→liq	1449±3	3.440	2.37

Selected Properties of Americium Metal

Property	Value
Atomic number	95
Density	13.671 g/cm^{-3}
High-pressure structures	0-5 GPa, dhcp 5-10 GPa, fcc 10-15 Gpa, double body-centered monoclinic >15 Gpa, orthorhombic –uranium or monoclinic (α-uranium alloys)
Melting point	1176, 1173, or 1170^{0}C
Boiling point	2067^{0}C or 2284^{0}C
Vapor pressure	log(p/atm) = (6.5780±.046)-(14315±55)/T at 990-1358K
Heat of vaporization at boiling point	230.2KJmol^{-1}(calc.)
Entropy of vaporization at boiling point	100.8 J K^{-1}mol^{-1}
Heat of fusion	14.4KJmol^{-1}

For the α–phase we estimate the following value:

$$C_p(Am,\alpha) = 5.294 + (2.883*10^{-3})T + (0.272*10^{-6})T^2$$

For the β–phase we estimate the following value:

$$C_p(Am,\beta) = 5.196 + (1.964*10^{-3})T + (7.38*10^{-7})T^2$$

Thermodynamic Quantities For Americium Ions:

Ion	ΔH_f^0(298K), KCal·mol^{-1}	ΔG_f^0(298K) KCal·mol^{-1}	S^0(298K) Cal·K^1·mol^{-1}	Hydration enthalpy and entropy	
				$-\Delta H_h$, KCal·mol^{-1}	$-S_h$, Cal·K^1·mol^{-1}
Am^{3+}·aq	147.4±0.3	143.2±0.9	48±3	832	91.8
Am^{4+}·aq	103.4±2.6	89.2±2.4	97±5	1635	128
AmO_2^{+}·aq	192.4±1.1	177.7±1.3	3±2		
AmO_2^{2+}·aq	155.8±0.5	141.0±0.8	19±2		

Decay heat from ^{241}Am:

- $t_{1/2}$ = 432 years
- $\lambda = \frac{\ln 2}{t_{\frac{1}{2}}} = \frac{0.693}{432 \times 365 \times 24 \times 3600} \cong 0.51 \times 10^{-10}\ s^{-1}$

$$\dot{R} = \lambda NE = (0.51 \times 10^{-10}\ \frac{dis}{s}) \cdot (\frac{13.6 \frac{g}{cm^3} \times 6.023 \times 10^{23}\ mol^{-1}}{241\ \frac{g}{mold}}) \cdot (4\ \frac{\mu ev}{dis})$$

$$= (69.36 \times 10^{11}\ \frac{\mu ev}{s - cm^{-3}}) \cdot (1.602 \times 10^{-7}\ \frac{J}{\mu ev})$$

$$= 1.11 \times 10^{6}\ \frac{W}{cm^3}$$