Contents lists available at ScienceDirect



journal homepage: www.elsevier.com/locate/jnucmat

Metallic fast reactor fuel fabrication for the global nuclear energy partnership

Douglas E. Burkes*, Randall S. Fielding, Douglas L. Porter

Idaho National Laboratory, Nuclear Fuels and Materials Division, P.O. Box 1625, Idaho Falls, ID 83415, USA

ABSTRACT

Fast reactors are once again being considered for nuclear power generation, in addition to transmutation of long-lived fission products resident in spent nuclear fuels. This re-consideration follows with intense developmental programs for both fuel and reactor design. One of the two leading candidates for next generation fast reactor fuel is metal alloys, resulting primarily from the successes achieved in the 1960s to early 1990s with both the experimental breeding reactor-II and the fast flux test facility. The goal of the current program is to develop and qualify a nuclear fuel system that performs all of the functions of a conventional, fast-spectrum nuclear fuel while destroying recycled actinides, thereby closing the nuclear fuel cycle. In order to meet this goal, the program must develop efficient and safe fuel fabrication processes designed for remote operation. This paper provides an overview of advanced casting processes how straightforward process parameter modification can mitigate the loss of volatile minor actinides in the metal alloy melt.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Fast reactors are being considered as transmutation systems for long-lived minor actinides resident in spent nuclear fuels as well as in nuclear power generation, as evidenced by recent US Department of Energy programs [1–4]. These considerations are spurred on by several global factors looming in the very near future. These factors are (1) consequences of the greenhouse effect that may produce a carbon tax, (2) production of hydrogen and hydrogen-rich fuel cells to produce energy for transportation, (3) increased demand for potable and irrigation water, (4) proliferation concerns associated with separated plutonium, and (5) determination of an appropriate spent-fuel strategy and repository.

Fast reactors are poised to address these factors since fast reactor fuels provide adequate long-term management of plutonium and minor actinides, minimizing proliferation risks and waste depository requirements, while generating a respectable amount of heat for energy, hydrogen, or water production. One of the two leading candidates for next generation fast reactor fuels is metal alloys, resulting from the notable successes obtained from driver fuel utilization and/or testing in the 1960s to early 1990s in both the experimental breeding reactor-II (EBR-II) and the fast flux test facility (FFTF). Metal fuel has the advantage of simple fabrication, high thermal conductivity, high fissile and fertile density capability, and small Doppler reactivity feedback [5]. Furthermore, metallic fuel can be simply recycled using either melt refining or an electrorefining process. Both processes have shown the ability to effectively remove a majority of fission products and return uranium, plutonium, and minor actinides to the reactor, leading to an economical and proliferation-resistant reprocessing scheme [6,7]. Metal fuel does have some disadvantages when compared to alternative fast reactor fuels (e.g., oxides), such as a low melting temperature and higher propensity for fuel-cladding chemical interaction. A number of experimental fuel tests, as well as qualified designs of driver fuel, are summarized in Table 1.

The varied designs progressed as performance of the fuel was better understood. The design was altered to improve the useful burnup (BU) of the fuel [8]. For example, the Mk-IA design did not provide for fuel swelling, axial or radial, and had very little plenum volume to accommodate fission gas released from the fuel. As a result, the fuel pins failed from stress rupture of the cladding at relatively low fuel BU, limiting the lifetime of a given driver fuel element.

Mk-II designs allowed for fuel swelling by a reduction in the areal smeared density, and also by providing an adequate plenum volume to prevent plenum gas pressure from causing cladding failure to >10 at.% BU. However, the Mk-II series used solution-annealed (SA) 316 stainless steel (SS) cladding that had very little resistance to void swelling.

The Mk-III and -IIIA designs used more swelling-resistant cladding (cold-worked D9 SS and 316 SS) and had increased plenum volumes. Test assemblies demonstrated that the fuel could easily exceed 15 at.% BU without breach of cladding. Unfortunately, the fuel assembly hardware could not be used to 15 at.% BU because the equivalent neutron dose produced too much hexagonal fuel duct expansion to permit fuel handling through the EBR-II storage basket. As a result, the fuel was limited to 10 at.% BU. Assembly



^{*} Corresponding author. Tel.: +1 208 533 7984; fax: +1 208 533 7863. *E-mail address:* Douglas.Burkes@inl.gov (D.E. Burkes).

^{0022-3115/\$ -} see front matter @ 2009 Elsevier B.V. All rights reserved. doi:10.1016/j.jnucmat.2009.03.009

	Table 1					
Selected design parameters (nominal) for EBR-II and FFTF metal driver fuel elements [9].						
		NA 1 X/ XA	N/ 1 W -			

Design	Mark-I/-IA	Mark-II series	Mark-III series	Mark-IV, V ^b	Series IIIB
Reactor	EBR-II	EBR-II	EBR-II	EBR-II	FFTF
Fuel alloy (wt%)	U–5Fs ^a	U–5Fs	U-10Zr	U-10Zr	U-10Zr
Slug diameter (mm)	3.66	3.30	4.39	4.27	4.97
Areal smeared density (%) ^c	85	75	75	75	75
Plenum-to-fuel volume ratio	0.2	0.7-0.1	1.4	1.4	1.5
Cladding material	304L	SA316	CW316, CWD9	HT9	HT9
Maximum burnup achieved (at.%)	2.6	8.9	10	N/A	14.3

^a Fs represents an equilibrium mixture of noble metal fission products, primarily comprised of Mo and Ru with some Rh, Pd and Zr.

^b Conversion to the Mark-V/-VA fuel types was never begun before EBR-II was terminally shut-down in 1994.

^c Smeared density is defined here as the ratio of the cross-sectional area of the as-fabricated fuel slug to the cross-sectional area defined by the cladding inner diameter.

hardware made from the ferritic/martensitic steel HT9 was to be included in future designs to allow even greater fuel utilization. However, these tests never came to fruition due to the terminal shut-down of the EBR-II facility in 1994.

The series IIIB, to be used as driver fuel in FFTF, used HT9 assembly hardware and was tested in six full assemblies to 11–16 at.% BU without any fuel pin breaches. Some of these assemblies were designed to produce peak fuel cladding temperatures (PCT) near 650 °C, often considered as a limit for conventional HT9 (620 °C PCT without uncertainties) due to lower stress rupture properties at higher temperatures [10].

Through the course of these design evolutions, the fabrication techniques also evolved with goals to allow remote fabrication, reduce waste, and simplify the process. The specifications were also relaxed as the fuel performance testing demonstrated the 'forgiving nature' of metal fuel in terms of impurities, dimensional tolerances, and thermal bonding imperfections.

2. Designing for global nuclear energy partnership (GNEP)

Development of fuel alloys for use in the transmutation of minor actinides such as americium, neptunium, and curium is one of the top goals of the global nuclear energy partnership (GNEP) program. Transmutation fuels for use in fast reactors must behave in a benign manner during core off-normal events, maintain integrity up to a high BU, lend themselves to low-loss recycling processes, and be easily fabricated with minimal material loss in a remote handling environment. For transmutation fuels (containing the minor actinides), these requirements are essential for the successful function of the fuel. This paper will focus on the last requirement for transmutation fuels: simplicity of fabrication with minimal material loss in a remote handling environment. The addition of minor actinides (MAs) Am, Np, and Cm as constituents in the fuel, as well as the potential for carryover of substantial guantities of rare earth fission products (such as cerium and neodymium), create a special set of challenges to future fuel design and fabrication not necessarily experienced in the EBR-II and FFTF irradiation campaigns. All of these alloying elements create potential performance issues, such as affecting fuel/cladding chemical interaction, radial composition redistribution, and melting temperature.

The alloying elements can also significantly influence fabrication processes. Some, such as Am, possess high vapor pressures and are therefore susceptible to creating a process loss during a casting operation. Not only is retention of Am important for successful transmutation of MAs in a fast reactor, but any material lost must be efficiently recovered in a waste stream and subsequently dealt with, increasing the overall facility cost and safety requirement. This is especially challenging in a remote fabrication environment, where all operations must either be automated or conducted with manipulators. Fig. 1 compares the vapor pressure of Am to other components of metal fuel.



Fig. 1. Vapor pressure of americium, plutonium, neptunium, and uranium as a function of temperature. The vapor pressure of Am is three orders of magnitude greater than that of Pu, which is four orders of magnitude greater than that of U [12–18].

In addition to potential material losses, the freezing range of a given alloy tends to increase as the number of alloying components becomes more complex, limiting the types of casting processes that may be used effectively. For these two reasons, the fabrication processes that have traditionally been used are being re-examined. Casting processes that do not require application of a vacuum are being designed with minimization of thermal gradients within the crucible and furnace. Calculations have predicted that loss of materials with high vapor pressure is virtually eliminated with simple modifications to existing injection casting designs and fabrication parameters [11].

Americium is a relatively minor component in proposed transmutation fuel alloys, approximately only 5 wt%. However, the vapor pressure (p_{Am}) of Am is so large (Fig. 1) that the partial pressure $(P_{Am} = c_{Am} \cdot p_{Am})$ in the alloy will dominate the gas mixture produced by the alloy components above the fuel solidus/liquidus during fabrication, where c_{Am} is the concentration of Am as a solute in the molten fuel. This leads to great concern that the Am could be lost as a gas during fabrication of fuel slugs, thus requiring additional measures to condense and recover the lost gas so the material can be returned to the feedstock stream. The potential loss of Am during high-temperature fabrication can be represented by a simple evaporation problem. Evaporation of the Am gas is not only accelerated by the high vapor pressure, but additionally requires diffusion away from the area above the heated alloy in either its solid or liquid state, depending upon the time in process. Diffusion of gas away from the interface can be influenced by a number of easily controllable parameters that either enhance or deter the Am loss from the solid or liquid. These parameters include time, temperature, pressure (and gas composition), and concentration. Furthermore, careful selection and design of an appropriate fabrication process for metal fuel alloys can minimize potential material losses. What follows is a review of processes previously investigated for fabrication of metal fuel alloys, and a discussion of a simple gaseous diffusion calculation that will analyze the influence of processing parameters using an injection casting process.

2.1. Review of advanced metal fuel fabrication technologies

In previous fuel fabrication campaigns for EBR-II, injection casting was the most effective technique for metal fuel fabrication. The process involves placing the feedstock metal into yttria-coated graphite crucibles, followed by melting in a high-frequency powered pressure/vacuum induction furnace at approximately 1500 °C [19,20]. Placement of feedstock into the crucible charge is important to aid the alloying process and minimize volatile component loss. Components with a low melting temperature, such as Pu, are loaded into the bottom of the crucible, followed by zirconium and finally uranium. Thus, the lower melting temperature U will flow over and consume the higher temperature Zr, and reduce the ability of higher vapor pressure constituents to escape into the vacuum. The outside of the crucible is typically wrapped in zirconia felt to insulate the crucible from the induction coil and to prevent significant heat loss. The induction coil must be made from a solid, oxygen-free, high-conductivity copper in order to reduce resistive heating and serve as a heat sink, since water cooling is typically not permitted inside the furnace shell. The furnace is evacuated before each injection casting run, followed by lowering of the molds below the surface of the melt. Once the molds have pre-heated for several seconds, the furnace is rapidly pressurized with argon gas, forcing the molten alloy into the molds. The molds themselves are the most problematic element of the injection casting process, since they must also be designed for a relatively high melting temperature. Glasses, such as silica and Vycor, were typically used as mold materials. However, as the mold must be broken from the cast fuel slug, the glass is not re-usable, and therefore not cost-efficient. Most significantly, the by-product mold shards created a highly undesirable additional waste recovery stream.

Centrifugal casting was used to fabricate U-2 wt% Zr alloy fuel for EBR-I [21,22]. The fuel slugs for EBR-I were significantly larger in diameter than for EBR-II (9.8 mm compared to 3.3-4.4 mm), as they would be for a commercial fabrication line, similar to those irradiated in FFTF (series IIIB). EBR-II required fabrication of longer and smaller-diameter fuel slugs. The centrifugal casting technique was found to be effective in casting rod-type fuel slugs with a surface-to-volume ratio of 26:1 and a length-to-diameter ratio of 59:1 composed of uranium and U-2 wt% Zr alloy. The success rate and surface finish of the castings was improved by using mold materials made of brass, copper, or silver. In addition, the re-design of the melt distributor had a profound impact on the ability of the process to cast longer fuel slugs with smaller diameters. In effect, centrifugal casting could potentially be used to fabricate fuel slugs with dimensions typical of a commercial fast reactor, while also minimizing volatility issues. However, the process has always been considered somewhat complicated and time consuming. The number and type of manipulations required to assemble and disassemble the furnace and molds are significant, and there are concerns over the relatively low production potential per machine, compared with other fabrication processes [23].

Continuous casting was another advanced fabrication method investigated, mainly because it eliminated the use of molds and produced fuel slugs with more uniform properties, since the solidification front was quasi-steady-state [24]. Bronze rods up to 91 cm long were cast with a diameter within ±0.06 mm and had a smooth, uniform surface finish. Although a continuous caster was built and staged to fabricate U–10 wt% Zr, tests were never conducted. It was generally believed that casting of bronze was a worst-case scenario compared to the U–10 wt% Zr, based on more favorable thermal and mechanical characteristics of the fuel alloy.

However, the cooling rate of cast material is essential in the success of the continuous casting method. Cooling becomes much more difficult with alloys containing the minor actinides, since the freezing range increases significantly. For example, the U–19Pu–10Zr alloy liquidus is at 1300 °C while the solidus is at 1080 °C, a 220 °C freezing range [25]. The wide freezing range can lead to micro-shrinkage effects and loss of process control during casting. Furthermore, pulling of the cast must be properly aligned to avoid any asymmetric variations in the rod diameter, increasing the complexity of the unit for remote operation. Finally, if continuous casting was to be used, the process would need to be highly automated to minimize the extent of human interaction required for casting a significant number of fuel slugs.

Schematics of the injection casting, centrifugal casting, and continuous casting processes are provided in Fig. 2. More recently, additional modifications to traditional fabrication processes have also been investigated. These include cold crucible casting and a bottom pour process. Each of these processes face their own set of challenges, both with implementation and operation in a remote environment. However, as it will be demonstrated, these newer processes are being developed to retain Am, based upon calculating gaseous Am loss.

2.2. The gaseous diffusion calculation

A simple gaseous diffusion calculation can be conducted to determine the flux of Am gas away from the alloyed melt as represented by the following equation:

$$N_{Am} = D_{Am} \cdot \frac{\{C_{Am}(x=0) - C_{Am}(x=L)\}}{L}.$$
 (1)

For Eq. (1), N_{Am} is the molar flux of Am, and D_{Am} is the diffusion coefficient of Am as a function of temperature, pressure, and cover gas. C_{Am} is the concentration of Am in the space surrounding the alloy melt at the surface (x = 0) and at some distance away from the surface (x = L, the heat sink, wall, etc.). Ultimately, the molar flux can be controlled by changing the concentration gradient (C_{Am}) or by altering the diffusion coefficient (D_{Am}).

The diffusion coefficient can be altered by influencing two operational parameters: temperature or pressure (including gas composition). No information exists on the diffusion coefficient of Am. However, the diffusion behavior can be extrapolated from known diffusion coefficients for species with similar mass as an initial starting point. For example, data is available for diffusivity of mercury in one atmosphere of argon and in some other noble gases [26]. Mercury has an atomic mass of 200 g mol⁻¹ while Am has an atomic mass of 243 g mol⁻¹. The diffusion coefficients for Hg must be extrapolated well above the existing data regime to be in a similar range for transmutation fuel processing temperatures, since the diffusion coefficients were measured at much lower temperatures. An estimation of the influence of pressure on the diffusion coefficient may be made by assuming a constant value of the diffusion coefficient multiplied by the system pressure [27]. Thus, by assuming a twofold increase in pressure, the diffusion coefficient would subsequently be halved.

The cover gas used during fabrication will also have a strong influence upon the diffusion coefficient of the Am gas. If the cover gas atomic/molecular species is larger and heavier than the Am gas, then diffusion becomes much more challenging for solute gas (Am) species. This is best demonstrated by extrapolation of a metal with similar behavior. For example, the diffusion coefficient of Zn gas at 700 °C in atmospheric helium is four times that of Zn



Fig. 2. Schematics of the injection casting (A), centrifugal casting (B), and continuous casting (C) processes investigated to fabricate metallic fast reactor fuel.

gas in Ar under the same conditions [26]. Similarly, the diffusion coefficient of Hg gas at 200 $^{\circ}$ C in He is more than four times that of Hg gas in Ar under the same conditions [26]. This is illustrated graphically in Fig. 3.

A reduction in the concentration gradient could reduce the loss of Am as it relates to the molar flux and diffusion of Am gas represented by Eq. (1). In other words, a minimal evaporation loss could be realized by using a closed system with a relatively small volume, as opposed to an open system or a large processing volume. The most obvious approach for capture of volatile Am gas is employment of a cold trap position near the alloy processing zone. The Am would condense in the cold trap, be dissolved out of the cold trap frit, and reintroduced into the feedstock processing stream after conversion back to metal. The most severe limitation with this approach is that the cold trap increases, and actually maximizes the concentration gradient. Therefore, the Am loss from the alloy melt is increased by use of a cold trap. If the control of temperature and pressure are not effective in controlling the diffusion coefficient, then perhaps a cold trap could be employed as a solution. However, values based on extrapolated diffusion coefficients seem reasonable and suggest that Am recycling need not be pursued as the primary, most logical method.

As an example, the following system (based upon injection casting) is used: a melt containing 5 wt% Am is held at 1750 K for 5 min with heated crucible walls, enabling the crucible lid as a perfect heat sink. The vapor pressure of Am, and thus the partial pressure,

Fig. 3. Diffusion coefficient of mercury and zinc under both argon and helium cover gas. The diffusion coefficients of Hg and Zn under He are much greater than those under Ar. After Ref. [26].

can be determined as explained above, e.g., 488 and 18.1 Pa, respectively. The diffusion coefficient of Am at one atmosphere is extrapolated to 1750 K based on the diffusion coefficient of Hg in Ar, mainly as a result of the low melting temperature of Hg. The Am diffusion coefficient at Ar pressure is then determined. The Am partial pressure is used to determine the Am concentration at the melt surface, assuming the concentration at x = L is zero since the lid is a perfect heat sink. Based on Eq. (1), the molar flux is determined and a percent Am loss rate calculated. Depending on the hold time, the total percentage of Am loss can be computed. Two separate cases were investigated: injection casting under modest vacuum (670 Pa), and injection casting with a slight overpressure of 30 kPa (0.3 atm). Calculations for these two cases are summarized in Table 2. With a modest vacuum of 670 Pa, a relative loss of 0.3% of the Am in the melt would occur. The retention of 99.7% of the Am might not be acceptable for a fuel fabrication operation, especially one designed for remote operation. Relative Am loss could be reduced to only 0.006% by altering the D_{Am} with increased pressure, e.g., 30000 Pa (~0.3 atm), e.g., Case II. This pressure would still theoretically permit filling of the quartz molds (i.e., not a significant loss in vacuum, while maintaining a minor amount of Am loss as a gas). According to Henry's law, the modest increase in pressure will increase the amount of Ar in solution with the molten fuel. However, Ar dissolution in molten U should be negligible compared to fission gas dissolution, e.g., Xe/Kr, which is on the order of 1% per atomic percent burnup of fuel.

A second example involves altering the concentration gradient by raising the temperature of any potential heat sink to create a positive effect. The most probable heat sinks are cold traps, as mentioned above, but also the crucible wall and cover lid. Americium will deposit at the sink if the example is considered an ideal system. The Am will then have a partial pressure equal to the vapor pressure at the sink surface. Recall that at the melt surface, the partial pressure is reduced because of the 5 wt% Am concentration in the melt (first example, above). If the sink temperature is lower

Table 2
Summary of gaseous diffusion calculations for Am loss in an injection casting process
conducted under a modest vacuum (Case I) and a slight Ar overpressure (Case II).

Case	Ι	II
Ar pressure (Pa)	670	30 000
Am diffusion coefficient at Ar pressure (cm ² s ⁻¹)	219	4.89
Am loss (%)	0.3	0.007

than the melt temperature and the Am vapor pressure generated is equal to the pressure just above the melt, then there is no driving force for diffusion away from the melt surface. Because pure Am at 1440 K has a vapor pressure of 18 Pa, increasing the temperature of the crucible walls and lid to 1440 K achieves an equilibrium of the vapor pressure of Am and partial pressure of Am in the melt. Therefore, no continuous and appreciable diffusion of Am from the melt surface will occur (theoretically). While this is a simple calculation, it demonstrates the likelihood that equipment design and control of process parameters can be used to effectively control Am loss, and recovery of Am streams need not be the only solution for the Am retention problem. Obviously experimental 'check points' or benchmarks are needed to prove Am behavior with such modifications.

2.3. On the use of surrogate systems

One of the most challenging aspects of any research and development program is minimizing the amount of experimentation required. Of particular importance in the nuclear field are the challenges of minimizing the waste generated, the scale of the experiments, and the amount of material required upfront - all relating to time and cost. Use of surrogate materials and systems are common when working with nuclear materials. Surrogates, by definition, are materials or systems that function properly, accurately, and with known transitions in place of another. As described above, one of the most significant challenges with metal transmutation fuel alloy fabrication is dealing with Am volatility. Fig. 4 provides examples in the similarity of vapor pressure between proposed surrogate metals and that of Am. There are many potential candidates for surrogates of metallic Am, of which manganese is the most widely used and discussed. Trybus investigated the use of a U-Mn-Zr alloy system to simulate the loss of Mn [Am] during an injecting casting process [28]. The scope of the experiment was not sufficient to include and test hypotheses related to minimization of Mn loss from the melt. However, based on the simple gaseous diffusion calculation described in this paper, similar results can be obtained by substituting Mn for Am and determining the loss from a molten pool of a Mn-bearing alloy. Similar limitations exist on available diffusion coefficient data on Mn in Ar as was identified for Am, but the information for Zn in Ar can be extrapolated to explain this behavior since Zn has an atomic weight of 65 g mol⁻¹ and Mn has an atomic weight of 55 g mol⁻¹. Measured diffusion coefficients of both Hg and Zn in

Fig. 4. Vapor pressure of americium metal and three proposed surrogate metals: manganese, silver, and praseodymium. The vapor pressures of the metals are all on the same order of magnitude with one another [18,29,30].

one atmosphere of either Ar or He gas as a function of temperature are provided in Fig. 3.

Based on the extrapolated diffusion coefficient of Zn in Ar. the same analysis can be conducted using the gaseous diffusion calculation by substituting Mn for Am. Thus, 1 wt% Mn in the melt will be the same as 5 wt% Am in a U-Pu-Zr-MA melt, based on atomic percent conversions (i.e., both are equivalent to 3.7 at.%). An Ar pressure of 670 Pa results in a 3% loss from the melt of the original 1 wt% Mn under the same processing temperature, volume, and hold time. A loss from the melt of 0.07% of Mn results when the pressure is increased to 30 kPa (note that 1 atm = 101.3 kPa) under the same processing conditions. For these examples, the calculation is constrained to a case where the crucible walls are heated to approximately 1500 K to avoid Mn collection. However, the crucible cover is not heated, so it serves as a perfect heat sink. If the lid were also heated to 1500 K, the only loss would result from the partial pressure saturating the gaseous area above the melt surface, as discussed previously. Based on these numbers, the Mn surrogate provides loss values that are an order of magnitude higher than those predicted for Am. Obviously, Mn offers an attractive option in terms of its conservatively higher vapor pressure and comparable atomic mass to Zn for extrapolation of diffusion coefficients, but care must be exercised in terms of a surrogate for Am loss during processing (such as from a melt). Furthermore, the differences in electronic structure, thermal, and phase behavior must not be overlooked for surrogate work, but discussions of these are well outside the scope of this paper.

Additional metals, such as silver and praseodymium have also been proposed as viable surrogates for Am based on vapor pressure. Once again, there is no solid experimental data for diffusion coefficients of these two metals. However, a mathematical expression for diffusion coefficients of these metals, along with a host of others, has been proposed with reasonable certainty [31]. The expression is represented by the following equation:

$$D = D_0 \left(\frac{P}{P_0}\right) \left(\frac{T_0}{T}\right)^n. \tag{2}$$

For Ag, $D_0 = 7.9$ and n = 1.89, and for Pr $D_0 = 7.4$ and n = 1.86. The ratio of pressures is taken as unity, since this will be accounted for in the gaseous diffusion calculation. Thus, at a temperature of 1750 K and assuming $T_0 = 273$ K, the diffusion coefficients are 2.99×10^6 and 2.34×10^6 m 2 s $^{-1}$ for Ag and Pr, respectively. Entering the diffusion coefficient of Ag into the calculation, altering the mass of Ag in the melt to correspond to that of Am, and using the conditions established as before for Am and Mn, the loss at 670 Pa is 1.4%, and at 30 kPa is 0.03%. Similarly for Pr, the loss is 0.86% at 670 Pa and 0.02% at 30 kPa. Thus, the correlation in percentage of mass loss to Am is better for Ag and Pr (in that order), than for Mn, but still an order of magnitude lower than that expected for Am. Furthermore, Ag and Pr face an additional set of challenges associated with their use as a surrogate material. Both Ag and Pr have very low solubility if uranium is used as the base, and a two-phase liquid(1)-liquid(2) molten regime, again with low solubility for each other. The partial pressure would then be nearly equivalent to the vapor pressure. This factor must always be considered when selecting an appropriate surrogate material; otherwise, results can be misinterpreted or poorly correlated with actual behavior.

3. Conclusions

The United States has a large existing database relating to the fabrication and performance of metal fuel alloys. With the emergence of advanced nuclear power generation, advanced nuclear fuels and fuel cycles are necessary. One such example is the transmutation of minor actinides in fast reactors while generating modest amounts of energy. However, these novel concepts are accompanied by the requirement of novel modifications to traditional fuel fabrication methods and process parameters, such as injection casting. An overview of past fabrication designs and fabrication processes investigated with associated advantages and disadvantages has been provided. In addition, a simple gaseous diffusion calculation has been developed to show how the variation in operational parameters can improve the retention of volatile species in the fuel alloy with excellent success. Finally, a discussion of potential surrogate metals to minimize time, cost, and exposure associated with volatile minor actinides revealed that care must be taken in the selection of a surrogate.

Acknowledgement

Work supported by the US Department of Energy, Office of Nuclear Energy (NE), under DOE Idaho Operations Office Contract DE-AC07-05ID14517.

References

- [1] A Technology Roadmap for Generation IV Nuclear Energy Systems, Issued by the US DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, 2002, Available through the US Department of Energy.
- US Department of Energy Office of Nuclear Energy, Science and Technology, The US Generation IV Implementation Strategy, 2003.
- [3] US Department of Energy, Report to Congress on the Advanced Fuel Cycle Initiative, 2003.
- US Department of Energy, Press Release, Department of Energy Announces New Nuclear Initiative, 2006.
- [5] D. Mohr, L.K. Chang, E.E. Feldman, P.R. Betten, H.P. Planchon, Nucl. Eng. Des. 101 (1987) 45.
- [6] L. Burris, R.K. Steunenberg, W.E. Miller, in: AICHE Symposium Series No. 254, vol. 83, 1987, p. 135.
- [7] Y.I. Chang, Nucl. Technol. 88 (1989) 129.
- [8] L.C. Walters, B.R. Seidel, J.H. Kittel, Nucl. Technol. 65 (1984) 1.
- [9] C.E. Lahm, J.F. Koenig, R.G. Pahl, D.L. Porter, D.C. Crawford, J. Nucl. Mater. 204 (1993) 119
- [10] D.C. Crawford, D.L. Porter, S.L. Hayes, J. Nucl. Mater. 371 (1-3) (2007) 202.
- [11] Status of Design Development for the Metal Fuel Casting Process, GNEP-AFCF-FUEL-TD-RP-2007-000367, Idaho National Laboratory, 2007.
- [12] N.D. Erway, O.C. Simpson, J. Chem. Phys. 18 (1950) 953.
- [13] H.A. Eick, R.N.R. Mulford, J. Chem. Phys. 41 (1964) 1475.
- [14] S.C. Carniglia, B.B. Cunningham, J. Am. Chem. Soc. 77 (1955) 1502.
- [15] R.I. Ackermann, E.G. Rauh, I. Chem, Thermodyn, 7 (1975) 211.
- [16] R.N.R. Mulford, Thermodynamics, in: Proceedings of a Symposium in Vienna, 1965, vol. 1, IAEA, Vienna, 1965, p. 231.
- A. Pattoret, J. Drowart, S. Smoes, Thermodynamics of nuclear materials, in: Proceedings of a Symposium in Vienna, 1967, IAEA, Vienna, 1968, p. 613.
- [18] J.W. Ward, W. Muller, G.F. Kramer, Transplutonium elements, in: Proceedings of Fifth International Conference in Baden-Baden, 1975, European Institute for Transuranium Elements, Karlsruhe, 1975.
- [19] H.F. Jelinek, G.M. Iverson, Nucl. Sci. Eng. 12 (1962) 405.
 [20] C.W. Wilkes, G.L. Batte, D.B. Tracy, V. Griffiths, EBR-II Fuel Slug Casting Experience, ANL-IFR-73, Argonne National Laboratory, 1987.
- [21] A.B. Shuck, The Development of Equipment and Methods for Centrifugally Casting Reactor Fuel Slugs, ANL-5123, Argonne National Laboratory, 1953.
- [22] A.B. Shuck, The Manufacture of Enriched Uranium Fuel Slugs for the Experimental Breeder Reactor, ANL-4847, Argonne National Laboratory, 1963.
- F.L. Yaggee, A Report on Some Attempts to Cast Centrifugally Fuel Elements of Small Diameter, ANL-6177, Argonne National Laboratory, 1961
- [24] M.A. Shimko, T.J. Jasinski, Apparatus for the Continuous Casting of Metallic Nuclear Fuel, Phase II Final Report, TM-1491, Creare, 1988.
- [25] L. Leibowitz, E. Veleckis, R.A. Blomquist, A.D. Pelton, J. Nucl. Mater. 154 (1988) 145
- [26] P.J. Gardner, P. Pang, S.R. Preston, J. Chem, Eng. Data 36 (1991) 265.
- [27] B.E. Poling, J.M. Prausnitz, J. O'Connell, The Properties of Gases and Liquids, Diffusion Coefficients, 5th Ed., McGraw Hill, 2000 (Chapter 11).
- [28] C.L. Trybus, J. Nucl. Mater. 224 (1995) 305.
- [29] O. Kubaschewski, Materials Thermochemistry, 6th Ed., Pergamon, New York, 1993
- P.L. Woolf, G.R. Zellars, E. Foerster, J.P. Morris, Vapor Pressures of Liquid [30] Manganese and Liquid Silver, US Department of the Interior, Bureau of Mines, 1960
- [31] B.V. L'vov, V.G. Nikolaev, Zhu. Prik. Spektroskopii (Engl. Trans.) 46 (1987) 7.