Nuclear Engineering and Design 286 (2015) 1–17

Contents lists available at ScienceDirect

Nuclear Engineering and Design

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

Modeling constituent redistribution in U–Pu–Zr metallic fuel using the advanced fuel performance code BISON

J. Galloway[a], C. Unal[a,*], N. Carlson[a], D. Porter[b], S. Hayes[b]

[a] Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545, United States
[b] Idaho National Laboratory, 2525N. Fremont Avenue, P.O. Box 1625, Idaho Falls, ID 83415, United States

HIGHLIGHTS

- An improved constituent distribution formulation in metallic nuclear fuels.
- The new algorithm is implemented into the advanced fuel performance framework BISON.
- Experimental Breeder Reactor-II data, T179, DP16, T459 are reanalyzed.
- Phase dependent diffusion coefficients are improved.
- Most influential phase is gamma, followed by alpha and thirdly the beta phase.

ARTICLE INFO

Article history:
Received 26 February 2014
Received in revised form 11 January 2015
Accepted 26 January 2015

ABSTRACT

An improved robust formulation for constituent distribution in metallic nuclear fuels is developed and implemented into the advanced fuel performance framework BISON. The coupled thermal diffusion equations are solved simultaneously to reanalyze the constituent redistribution in post irradiation data from fuel tests performed in Experimental Breeder Reactor-II (EBR-II). Deficiencies observed in previously published formulation and numerical implementations are also improved. The present model corrects an inconsistency between the enthalpies of solution and the solubility limit curves of the phase diagram while also adding an artificial diffusion term when in the 2-phase regime that stabilizes the standard Galerkin finite element (FE) method used by BISON. An additional improvement is in the formulation of zirconium flux as it relates to the Soret term. With these new modifications, phase dependent diffusion coefficients are revaluated and compared with the previously recommended values.

The model validation included testing against experimental data from fuel pins T179, DP16 and T459, irradiated in EBR-II. A series of viable material properties for U–Pu–Zr based materials was determined through a sensitivity study, which resulted in three cases with differing parameters that showed strong agreement with one set of experimental data, rod T179. Subsequently a full-scale simulation of T179 was performed to reduce uncertainties, particularly relating to the temperature boundary condition for the fuel. In addition a new thermal conductivity model combining all available data covering 0–100% zirconium concentration and a zirconium concentration dependent linear heat rate solution derived from Monte Carlo N-Particle (MCNP) simulations were developed. An iterative calibration process was applied to obtain optimized diffusion coefficients for U–Pu–Zr metallic fuels. Optimized diffusion coefficients suggest relative improvements in comparison to previous reported values. The most influential or uncertain phase is found to be the gamma phase, followed by alpha phase, and thirdly the beta phase; indicating separate effect testing should concentrate on these phases.

1. Introduction

In the past, both metallic and oxide nuclear fuels were tested in fast reactor programs and performed well (Walters, 1999). The recent nuclear accident at Fukushima shifted the advanced fuels research focus to the development of accident tolerant fuels for light water as well as small modular reactor applications. The use of metallic fuels in these

Abbreviations: EBR-II, Experimental Breeder Reactor-II; FE, Galerkin finite element; LWR, light water reactor; M&S, modeling and simulation; MCNP, Monte Carlo N-Particle; US, United States.

* Corresponding author at: Los Alamos National Laboratory, P.O. Box 1663, Mail Stop F606, Los Alamos, NM 87545, United States. Tel.: +1 505 665 2539; fax: +1 505 665 5204.
E-mail address: cu@lanl.gov (C. Unal).

http://dx.doi.org/10.1016/j.nucengdes.2015.01.014
0029-5493/

new nuclear technology concepts has been gaining renewed attention.

Metallic fuels are attractive since they have higher thermal conductivity with a highly conductive gap (typically sodium) that enables the fuel to operate at lower temperatures with reduced stored energy. Additional benefits of metallic fuels include a more favorable neutron economy, higher fuel densities, and easier fabrication and reprocessing.

Metallic fuels tested previously typically consisted of a binary alloy of uranium and zirconium, or a ternary alloy of uranium, plutonium and zirconium. The addition of ~10% by weight zirconium to a uranium–plutonium alloy raises the solidus temperature of the fuel and prevents fuel cladding inter diffusion at temperatures typical of steady operating conditions (Walters, 1999). The addition of americium, neptunium, and curium to the traditional metallic fuel is one aspect considered for closing the fuel cycle. New metallic fuel concepts also include the use of uranium–molybdenum and uranium–palladium–zirconium based fuels.

Generally speaking, the thermo-mechanical behavior of a nuclear fuel pellet or rod involves a complex system of interdependent processes as a result of the high thermal-power densities and irradiation effects. All of the physics are driven by processes occurring at the microstructure level (i.e., at the grain or subgrain scale). The migration of porosity, fuel constituents, and fission products causes fuel restructuring in metal fuels. Irradiation induced effects such as fission product generation, as well as chemical interactions, change the material properties of the fuel and cladding. Microstructural characteristics and their irradiation induced evolution can also influence material properties. Fuel swelling, a result of both gaseous and solid fission product generation, and atomic displacements due to the neutron flux, have significant impacts on dimensional changes and mechanical properties of the materials. Swelled fuel causes mechanical interaction between fuel and cladding that can produce stresses and deformation in addition to the stress caused by internal pressure in the fuel. The nuclear (heat and irradiation) source drives many of these processes, which can vary greatly in thermal reactors. In addition, the chemical interactions between cladding and coolant significantly affect the heat transfer coefficient and mechanical strength of the cladding; these interactions can be driven by both local (nucleate boiling) and global (soluble metals) coolant flow conditions.

The behavior of metallic fuel undergoes various stages during irradiation. A detailed description of the metallic fuel behavior under irradiation is available in previously published papers (Walters, 1999; Carmack et al., 2009; Hofman et al., 1996; Karahan and Buongiorno, 2009). One important key design and modeling issue with metallic fuels has been found to be the constituent distribution; specifically zirconium redistribution (Walters, 1999; Carmack et al., 2009; Hofman et al., 1996; Karahan and Buongiorno, 2009; Kim et al., 2004, 2005, 2006, 2009; Ishida et al., 1993). Fuel constituent migration affects the fuel slug material properties, such as solidus and liquidus temperatures, thermal conductivity, along with mechanical properties such as the modulus of elasticity, fuel cladding eutectic reaction rate, and radial power density profile.

Zirconium redistribution in U–Zr and U–Pu–Zr based fuels is important for both fuel integrity and thermal limits. As zirconium redistributes, the uranium moves in the opposite direction and what was once a uniform isotopic concentration across the fuel rod becomes concentrated in zirconium, primarily at rod center and rod edge. This preferential accumulation at rod edge and rod center, with a corresponding decrease in the central region can affect local thermal limits (margin to melting temperature for example), radial power peaking, with potential enhances to lanthanide and actinide migration, all due to a significantly varying radial isotopic composition. A strong understanding of this process along with a robust method for predicting zirconium concentrations is vital to advanced metallic fuel designs, particularly those designs with minor actinide (MA) loadings, lanthanide loadings and new proposed metallic alloys.

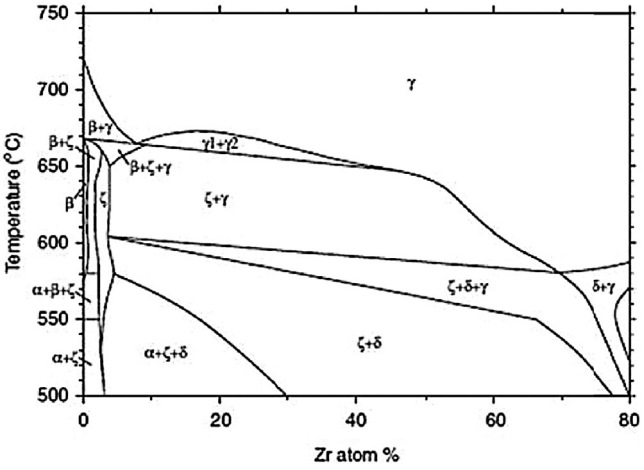

Fig. 1. Pseudo-binary (U–Pu)–Zr phase diagram, Pu content fixed at 19 wt% and 10%Zr from Refs. Kim et al. (2006), Ishida et al. (1993).

The prediction of zirconium redistribution is difficult for several reasons. First, fuel behavior under irradiation is a multi-physics, multi-scale problem that requires the coupling of neutronic solutions due to uranium migration which induces radial power peaking. Correspondingly, burnup effects in the fuel cause changes in both power and the thermo-mechanical-diffusion fuel behavior. Additionally there are thermal considerations primarily related to thermal conductivity; as zirconium migrates, the thermal conductivity changes inversely with zirconium concentration – an increase in zirconium concentration causing a decrease in thermal conductivity and vice versa. Fuel burnup increases porosity in the fuel, which degrades thermal conductivity. However, near the rod edge liquid sodium infiltration due to fuel cracking can occur, causing some recovery of the degraded conductivity. This porosity itself appears to be phase-dependent, particularly in the beta phase region of the fuel where porosity appears to be at a minimum and thermal conductivity degradation is expected to be lower. Along with neutronic and thermal–mechanical concerns, the phase properties of the fuel and our fundamental understanding of fuel properties and fuel behavior, particularly U–Pu–Zr based fuel, is not well known.

The microstructure of irradiated U–Pu–Zr fuel exhibits three distinct concentric zones, a zirconium-enriched central zone, a zirconium-depleted and uranium-enriched intermediate zone, and a zirconium-enriched zone on the outer periphery where the effective heat of transport drives the direction of zirconium migration atoms in different phases. Phase-dependent diffusion coefficients and heats of transport are not well known, with early evidence indicating the phase transition temperatures, experimentally derived from fresh fuel, may not be consistent with irradiated fuel. Final considerations are related to the phase diagram itself. Fig. 1 shows the experimentally derived phase diagram for U–19Pu–10Zr fuel from Refs. Kim et al. (2004, 2006). Noting the complexity and difficulty associated with programming logic for such a diagram, along with the lack of any material property information for several of the phases, a simplified version based on that used in Karahan and Buongiorno (2009) and Kim et al. (2004, 2006) was used in practice, shown here in Fig. 2.

There have been several published papers that analyze constituent distribution in metallic fuels mechanistically for U–Zr and U–Pu–Zr fuels (Karahan and Buongiorno, 2009; Kim et al., 2004, 2006; Hofman et al., 1996; Ishida et al., 1993). In the most recent