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THERMODYNAMIC AND KINETIC MODELING OF ADVANCED NUCLEAR FUELS - FINAL LDRD-ER REPORT

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Disclaimer

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Thermodynamic and Kinetic Modeling of Advanced Nuclear Fuels
Final Report of the LDRD-ER 10-ERD-059

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Fiscal Year: 2011
Tracking Code: 10-ERD-059
Title: Thermodynamic and Kinetic Modeling of Advanced Nuclear Fuels
PI: Patrice Erne Turchi

Project Description
WAIT FOR REVISED DATASHEET - PI SUBMITTED as a NEW SI (11-SI-003), but this is really a continuation of this project, 10-ERD-059). A key issue on the path to nuclear energy becoming an essential component of the U.S. clean energy strategy is complete burn of the nuclear fuel. We propose research that will enable high burn-up fuels by establishing the basic science for development and qualification of advanced nuclear fuel that will couple modern computational materials modeling, fabrication, and characterization capabilities with targeted performance-testing experiments using ion-beam facilities. This work will establish the scientific foundation and guide selection of the optimum fuel type for advanced reactor concepts.

Expected Results
We expect to experimentally quantify phase stability and kinetics of phase transformations, interdiffusion, microstructural evolution, micro-mechanical properties, and the influence of severe radiation environments on fuel performance and, by the end of the project, develop a validated model for advanced nuclear energy materials under extreme conditions of radiation, temperature, and evolving chemistry. We will provide a science-based path forward to an optimized inert matrix fuel, while contributing to the development of a validated nuclear-fuel database. We expect this project will establish LLNL's credibility within the nuclear energy community.

Mission Relevance
Our approach to development of the science of advanced nuclear energy fuels aligns well with the Laboratory's energy and national security missions. Development of both advanced fuel cycles and hybrid fusion-fission concepts is inexorably intertwined with the same scientific challenges. This research will extend LLNL capabilities and further enable actinide and high-energy-density science, high-performance computing and simulation, energy manipulation, and capabilities to develop materials on demand.

Accomplishments & Results
Three tasks were completed. First, enhance our modeling capabilities to assist experiments: 1) Extend the LLNL phase-field code to account for thermal transport and make it compatible with the thermodynamic and kinetic data as assessed in the CALPHAD approach, to predict microstructural changes in the presence of a thermal gradient; 2) Develop a search algorithm linked to the thermodynamic Thermo-Calc software for predicting optimum composition of a multi-component alloy with specific properties. Second, carry out ab initio

studies on actinide alloys including U-Mo, U-Ti, Np-Zr, X-Am (X=U,Np,Pu,Cm), and phase diagram assessments of Pu-Am, Np-Zr, and U-Ti (the last two in collaboration with TAMU). Third, position LLNL for funding opportunities. This includes: a proposal to DOE-NE on "Innovative Transmutation Fuels Concepts" (selected for funding, but allocation still pending); participation in an international "Thermodynamics of Advanced Fuels-International Database (TAF-ID)" project, initiated by CEA (France) submitted to OECD-NEA; and interaction with Dr. Savchenko and his team at the Bochvar Institute (VNIINM, Russia) to draft a proposal on the development of an innovative fuel.

Proposed Work/Summary Statement

This project enhanced our theoretical capabilities geared towards establishing the basic science of a high-throughput protocol for the development of advanced nuclear fuel that should couple modern computational materials modeling and simulation tools, fabrication and characterization capabilities, and targeted high throughput performance testing experiments. The successful conclusion of this ER project allowed us to upgrade state-of-the-art modeling codes, and apply these modeling tools to *ab initio* energetics and thermodynamic assessments of phase diagrams of various mixtures of actinide alloys, propose a tool for optimizing composition of complex alloys for specific properties, predict diffusion behavior in diffusion couples made of actinide and transition metals, include one new equation in the LLNL phase-field AMPE code, and predict microstructure evolution during alloy coring. In FY11, despite limited funding, the team also initiated an experimental activity, with collaboration from Texas A&M University by preparing samples of nuclear fuels in bulk forms and for diffusion couple studies and metallic matrices, and performing preliminary characterization.

Current text contains possible patentable material: N

ADC/DUSA:

Patents

Date Record of Invention (ROI) Filed ROI Number Date Patent Applied Date Patent Granted Patent Number Patent Title Inventors
No patents.

No Patents/ROIs:

Y

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*** Publications***

Authors	Pub. Year	Title	Journal/Conference	Vol.	Page	UCRL/LLNL No	Type
P.E.A. Turchi, A.I. Landa, V. Lordi, J.-L. Fattebert	2011	Thermodynamic and kinetic modeling of	EUROMAT 2011, Montpellier, France,	N/A	N/A	LLNL-ABS-470592	Conference/Meeting Abstract

		nuclear materials	Sept. 12-15, 2011				
A. Landa, P. Söderlind, P.E.A. Turchi, A.V. Ruban, L. Vitos	2011	Ab initio study of advanced metallic nuclear fuels for fast breeder reactors	EUROMAT 2011, Montpellier, France, Sept. 12-15, 2011	N/A	N/A	LLNL-ABS-466411	Conference/Meeting Abstract
J.-L. Fattebert, M.R. Dorr, M.E. Wickett, J.F. Belak, P.E.A. Turchi	2011	A numerical algorithm for the solution of a phase-field model of polycrystalline alloys	ICIAM2011, Vancouver, Canada, July 2011	N/A	N/A	LLNL-PRES-490951	Conference/Meeting Abstract
P.E.A. Turchi, A.I. Landa	2011	Actinide alloys and their challenges	CALPHAD XL, Rio de Janeiro, Brazil, May 22-27, 2011	N/A	N/A	LLNL-ABS-470593	Conference/Meeting Abstract
J.-L. Fattebert, M.R. Dorr, M.E. Wickett, J.F. Belak, P.E.A. Turchi	2011	A numerical algorithm for the solution of a phase-field model of polycrystalline alloys	APS March Meeting, Dallas, TX, March 2011	N/A	N/A	LLNL-PRES-474951	Conference/Meeting Abstract
Alexander Landa, Per Söderlind, Patrice E.A. Turchi	2011	Density-functional study of U-Mo alloys	APS March Meeting, Dallas, TX, March 2011	N/A	N/A	LLNL-ABS-462665	Conference/Meeting Abstract
V. Lordi, J.F. Belak, and P.E.A. Turchi	2011	Alloy optimization for metallic inert matrix nuclear fuels	TMS 2011, San Diego, CA, Feb 27-March 3, 2011	N/A	N/A	LLNL-ABS-444611	Conference/Meeting Abstract
P.E.A. Turchi, A.I. Landa, P. Söderlind, L. Kaufman	2011	Phase formation in actinide alloys: Why ab initio?	TMS 2011, San Diego, CA, Feb 27-March 3, 2011	N/A	N/A	LLNL-PRES-471468	Conference/Meeting Abstract
J.-L. Fattebert, M.R. Dorr, M.E. Wickett, J.F. Belak, P.E.A. Turchi	2011	A numerical algorithm for the solution of a phase-field model of polycrystalline alloys	SIAM Conference on Computational Science and Engineering, Reno, NV, February 2011	N/A	N/A	LLNL-POST-471650	Conference/Meeting Abstract
S. Bajaj, R. Arroyave, A. Landa, P. Turchi	2011	CALPHAD and DFT assessment of metallic alloy fuel materials	TMS 2011 140th Annual Meeting & Exhibition, San Diego, CA, February 2011	N/A	N/A	LLNL-ABS-442918	Conference/Meeting Abstract
P.E.A. Turchi, A.I. Landa, P. Söderlind, L. Kaufman	2010	Thermodynamics of metallic nuclear fuels - A current status	Nuclear Materials 2010 Conference, Karlsruhe, Germany, October 2010	N/A	N/A	LLNL-ABS-429152	Conference/Meeting Abstract
A. Landa, P. Söderlind, P.E.A. Turchi	2010	Density-functional study of the U-Zr and U-Mo alloys	Nuclear Materials 2010 Conference, Karlsruhe, Germany, October 2010	N/A	N/A	LLNL-ABS-430211	Conference/Meeting Abstract
J.F. Belak, J-L. Fattebert, G.L. Hofman, Y.S. Kim, A.I. Landa, V. Lordi, P.A. Söderlind, P.E.A. Turchi, A.M. Yacout	2010	Phase Stability of metallic nuclear fuels: A multi-scale coupling of phase-field modeling with CALPHAD and first-principles	Fifth International Conference on Multi-scale Materials Modeling, MMM 2010, Freiburg, Germany, October 2010	N/A	N/A		Conference/Meeting Abstract
P.E.A. Turchi, Per Söderlind, Larry Kaufman	2011	Thermodynamic assessment of advanced nuclear fuel forms	Xth International Workshop on Fundamentals of Plutonium, Moscow, Russia, July 12-16, 2010	N/A	N/A	LLNL-PRES-438598	Conference/Meeting Abstract
J. Belak, J.-L. Fattebert, A. Landa, P. Söderlind, P. Zepeda-Ruiz, P. Turchi	2010	Modeling UZr metallic fuels: Coupling thermodynamics with microstructure	2010 ANS Annual Meeting and Embedded Topical Meetings, Transactions of the American Nuclear Society	N/A	N/A		Conference/Meeting Abstract
P.E.A. Turchi, A. Landa, P. Söderlind	2011	Thermodynamic assessment of the Am-Pu system with input from ab initio	Journal of Nuclear Materials	418	165-173	LLNL-JRNL-480181	Refereed Publication
A.I. Landa, P. Söderlind, P.E.A. Turchi	2011	Density-functional study of the U-Zr system	Journal of Alloys and Compounds	478	103-110	UCRL-JRNL-405060	Refereed Publication
A. Landa, P. Söderlind, P.E.A. Turchi, L. Vitos, O.E. Peil, A.V. Ruban	2011	Density functional study of bcc Pu-U, Pu-Np, Pu-Am, and Pu-Cm Alloys	Journal of Nuclear Materials	408	61-66	LLNL-JRNL-441353	Refereed Publication
A. Landa, P. Söderlind, and P. E. A. Turchi	2011	Density-functional study of U-Mo and U-Zr alloys	Journal of Nuclear Materials	414	132-137	LLNL-JRNL-461538	Refereed Publication

A.I. Landa, P. Söderlind, P.EA. Turchi, L. Vitos, A. Ruban	2009	Density-functional study of Zr-based actinide alloys	Journal of Nuclear Materials	385	68-71	LLNL-CONF-405029	Refereed Publication
A. Landa, P. Söderlind, P.E.A. Turchi, L. Vitos, A. Ruban	2009	Density-functional study of Zr-based actinide alloys: 2. U-Pu-Zr system	Journal of Nuclear Materials	393	141-145	LLNL-JRNL-410491	Refereed Publication

Awards

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Final Report:

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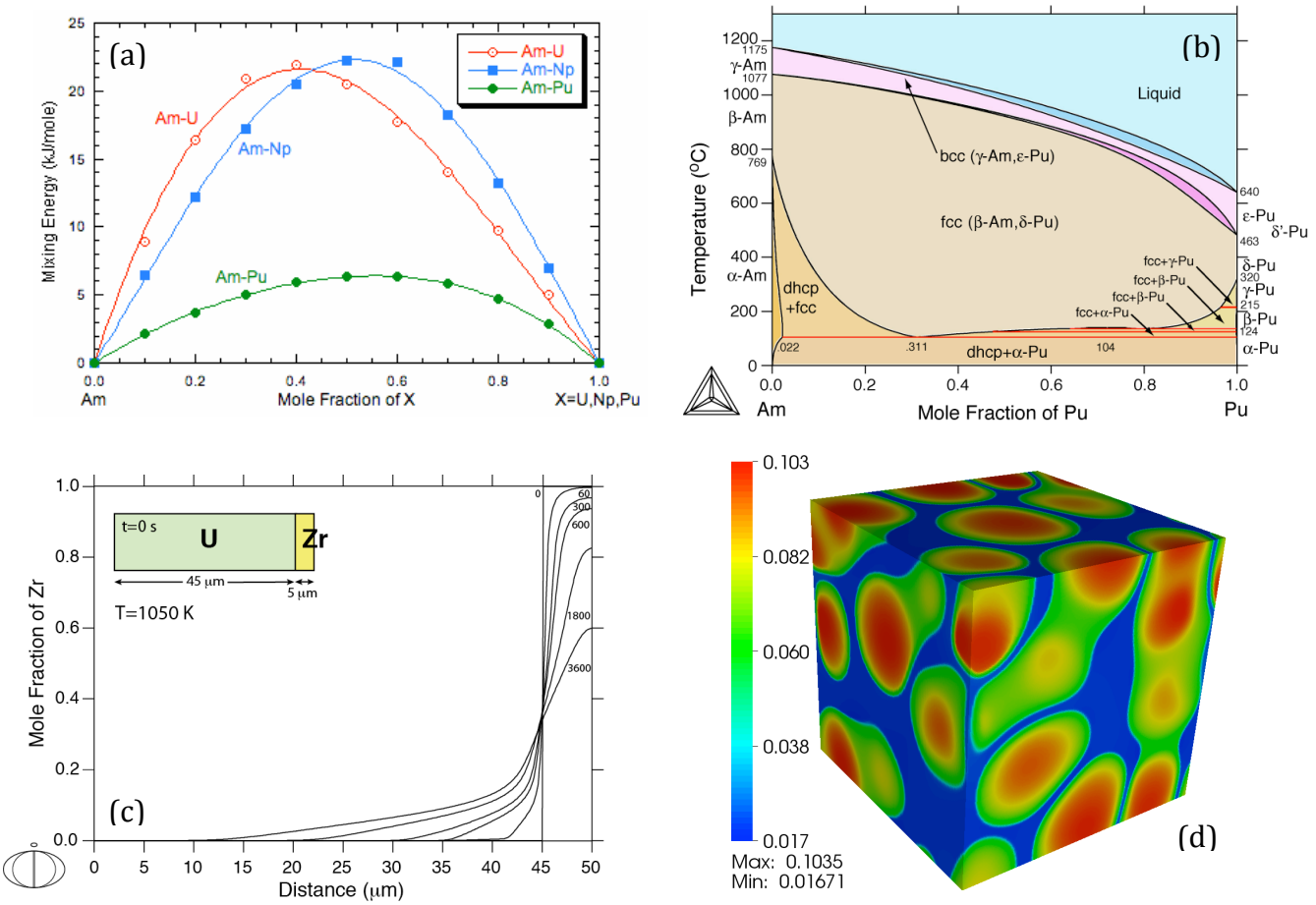

(a) Phase stability trends in bcc-based Am-X alloys (X=U,Np,Pu) from *ab initio* calculations. The positive heat of formation in all three cases indicates a tendency towards phase separation. (b) Thermodynamic assessment of the Am-Pu phase diagram based on the CALPHAD methodology with input energetic results from *ab initio* calculations. (c) Theoretical prediction of diffusion behavior (mole fraction of Zr versus distance) for a U/Zr diffusion couple (the initial sample geometry is represented on the top left corner, with 5 μm of Zr bonded to U) maintained at 1050 K, and for various times (60, 300, 600, 1800, and 3600 s). (d) Coring in alloys based on 3d-phase-field model simulations – the case of Pu-Ga involving the growth of the fcc phase (in green) from the bcc phase (in blue). Higher composition of gallium appears in the center of the fcc grains.