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Deep Burn Team



Deep Burn: Development of Transuranic Fuel for High-Temperature Helium-Cooled Reactors

Monthly Highlights

July 2010



TRISO-Coated Particle with Mixed Pu, Th Oxide
Kernel after High Pu Burnup

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Monthly Highlights for July 2010

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Acronyms and Abbreviations

ANL	Argonne National Laboratory
DB	Deep Burn; deep burn
DOE	Department of Energy
GA	General Atomics
GK	Green-Kubo method
HEPA	High Efficiency Particulate Air (filter)
HFIR	High Flux Isotope Reactor
HTR	high temperature helium-cooled reactor
IFEL	Irradiated Fuel Examination Laboratory (ORNL)
INEST	Institute for Nuclear Energy Science and Technology
INL	Idaho National Laboratory
LAMDA	Low Activation Materials Development and Analysis
LANL	Los Alamos National Laboratory
LOGOS	LOGOS Nuclear Systems
LWR	Light Water Reactor
METROX	metal recovery from oxide fuel
ORNL	Oak Ridge National Laboratory
PCEA	a candidate nuclear graphite manufactured by UCAR Carbon Co.
ppm	parts per million
REDC	Radiochemical Engineering Development Center (ORNL)
SEM	scanning electron microscopy
TAMU	Texas A & M University
TEM	transmission electron microscopy
TRISO	tri-structural isotropic
TRU	transuranic elements
UCB	University of California, Berkley
UNLV	University of Nevada, Las Vegas
UW-M	University of Wisconsin-Madison
XRD	X-ray Diffraction

1. Project Management and Planning

Program reporting (*ORNL*)

The DB Program Quarterly Progress Report for April - June 2010, ORNL/TM/2010/140, was distributed to program participants on August 4.

Archiving program records (*ORNL*)

The DB Program Quarterly Progress Report for April - June 2010 was posted on August 4 to the Deep Burn website, http://www.ms.ornl.gov/deep_burn/index.shtml. Program participants are requested to send reports, milestone documents and other pertinent documents to the webmaster, Shirley Shugart shugartsa@ornl.gov, for uploading to the website.

2. Core and Fuel Analysis

(No report for July)

3. Spent Fuel Management

(No report for July)

4. Fuel Cycle Integration of the HTR

(No report for July)

5. TRU HTR Fuel Modeling

5.1 Thermochemical Modeling

Thermochemical behavior (*ORNL*)

In anticipation of efforts on SiC matrix TRISO fuel for LWR applications, a review of the potential for SiC to react with zirconium in the clad was undertaken. Calculations of the reaction depth to form zirconium carbide were performed extrapolating high temperature-determined reaction rate constants to the lower temperature envisioned for LWR fuel, with the result that minimal interactions will occur.

5.2 Actinide and Fission Product Transport

Pd interactions with SiC (*ORNL*)

Work has been completed for investigating the mechanisms of silicon and carbon removal from SiC through palladium attack. The energy barrier for silicon removal is 1.52 eV. The barrier for carbon removal is 0.48 eV when a single Pd is involved and 0.04 eV if two Pd are involved. Thus, the rate limiting step has a barrier of 1.52 eV which is comparable to experimental work on palladium silicide formation at 500°C.

Fission product transport in PuO_{2-x} kernel (*LANL*)

Density functional theory calculations of the diffusion of palladium in PuO₂, focusing on Pd's movement on the oxygen sublattice, continue. The focus is on a large supercell, corresponding to a 1 atomic percent concentration of palladium atoms. Compared to a smaller supercell (corresponding to a 4 atomic percent concentration of palladium atoms) the larger cell provides a physically more realistic simulation of the fission product's diffusion.

Transport through coating layers (UCB)

Completing the designs of experiment to assess Ag diffusion within SiC layers. Preparing a letter report to describe the proposed experiments to assess Ag diffusion.

Diffusion mechanisms (UW-M)

Studies on diffusion of silver and cesium interstitials and vacancy clusters in ZrC have been performed. The defect formation energies of cesium and silver interstitials are 18.0 and 9.0 eV, respectively, which will greatly reduce their contribution to impurity diffusion. The cesium value is so high that no further study of this defect is warranted. However, the diffusion of a silver interstitials has been explored. The estimated effective diffusion coefficient of a silver interstitial is $3.0 \times 10^{-3} \exp[-7.7/kT]$ cm²/s. The diffusion mechanism of Ag_{Zr}-1Va_C point defect cluster has also been identified. The *ab initio* calculated activation energy for its effective diffusion is 12.6 eV, and the effective diffusion coefficient is predicted to be $1.9 \times 10^{-1} \exp[-12.6/kT]$ cm²/s. These results are being integrated with previous studies to develop a complete model for Ag diffusion in ZrC.

5.3 Radiation Damage and Properties

Thermal conductivity of SiC (ORNL)

The calculation of the thermal conductivity for a modeling cell of size 10x10x10 for additional concentrations of vacancies and voids in Si and SiC has been performed with the Green-Kubo (GK) method. The contribution of isotopic impurities to Si thermal conductivity has been investigated for sets of different masses. A sizable difference in relative change of thermal resistance produced by voids calculated by GK and by Debye-Callaway methods was observed. Both Molecular Dynamic and Debye-Callaway are being compared with experiment.

6. TRU HTR Fuel Qualification

6.1 TRU Kernel Development

Glove box installation in REDC (ORNL)

The paper entitled "Treatment techniques to prevent cracking of amorphous microspheres made by the internal gelation process," will be accepted by the *Journal of Nuclear Materials* after revisions.

Work of the installation of the internal gelation system for the fuel kernel fabrication in Building 7920 was completed. Tests of individual unit operations are underway.

6.2 Coating Development

Glove box installation in IFEL (ORNL)

The deep burn coating lab infrastructure design is continuing. A 60% design review meeting is set for the week of August 9th, 2010. Detailed design of the coating glove box is ongoing, as is procurement of the components of the new coating furnace. The ventilation concept for the coating glove box has again been revised. Currently the preferred concept calls for the glove box to operate with an inert internal atmosphere of argon while flammable gasses are present (during a coating run). The argon atmosphere will be achieved using a low-flow once-through supplied argon system. During the time when the flammable gasses are not present the supplied argon system will be valved off and air will be drawn at a low flow rate through the glove box. An oxygen sensor will be placed inside the glove box and will provide an additional condition that has to be met in order for the interlock valve on the process gas inlet line to open. Currently the anticipated interlock conditions include oxygen concentration, exhaust header flow, exhaust header temperature, and glove box internal pressure. The

pressure inside the glove box will be maintained slightly less than the surrounding room at all times, regardless of internal atmosphere. The process exhaust stream during coating operations will be routed from the furnace directly to the ventilation header where sufficient dilution will be achieved by the flow air within the header. This change significantly reduces the size of the inlet and outlet HEPA filters on the coating glove box and thereby simplifies the design of the system.

6.3 Characterization Development and Support

No activity

6.4 ZrC Properties and Handbook

Mechanical properties of zone-refined ZrC (ORNL)

A second revision of the ZrC properties handbook was completed and submitted for internal review.

Properties Measurement of ZrC (ORNL)

Discs of five different stoichiometric compositions of ZrC, each 3 mm thick, were prepared for high temperature strength testing. Approximately 60 discs of each composition were obtained using a slow speed diamond saw in the ORNL-LAMDA facility. Thickness measurements of random samples from batches of each composition yielded a value of 630 +/- 10 μm . Grinding of the samples to thin them to 250 +/- 10 μm thickness is in progress.

The modified push rod for the ultra high vacuum, high temperature furnace was received in the last week of July. The new design incorporated extended tungsten and molybdenum adaptors, replacing the original stainless steel design which was not suitable for temperatures exceeding 1400°C. Mechanical tests are scheduled to begin in late August.

Pd/ZrC interaction (UNLV)

Based on the results obtained for commercial ZrC reference materials, first studies of ORNL-prepared ZrC-coated particles were conducted at UNLV. Samples from batch ZrC17 and ZrC21 were studied, which differ in the preparation temperature and the carrier gas composition (ZrC17:1100°C and C_3H_6 ; ZrC21:1500°C and CH_4 ; see monthly report 09/2009). Both batches were grown with a C/Zr ratio of 1.2, but showed a different microstructure in the SEM and XRD studies reported in 9/09. XPS spectra show an enhanced Zr signal, a reduced C signal, and a similar O signal for the ZrC21 sample surface when compared with ZrC17. Consequently, the Zr/C ratio at the surface of the ZrC21 sample is also higher. Furthermore, both samples exhibit a Na signal at the surface, and, for ZrC17, Cl and Fe signals were also found.

Ag/ZrC interaction (UW-M)

Eight ingots of pure Zr were fabricated, which later will be alloyed into Zr-7at.%Ag solid solution ingots. The solid solution ingots will be used to construct [ZrC/Ag gas] constant source diffusion couples between a ZrC disk and an encapsulating Zr-Ag solid solution. A apparatus for diffusion annealing was also prepared.

Irradiation effects in ZrC (UW-M)

The cross-sectional TEM specimen of irradiated ZrC (1.1) was prepared and examined using TEM. The irradiation of ZrC at 1400 °C will be performed in August.

7. HTR Fuel Recycle

7.0 Recycle Processes (ORNL)

A presentation entitled “Deep Burn TRU Actinide Partitioning-Transmutation using Used Fuel Recycle and High Temperature Reactors (HTRs)” was prepared and presented at the Institute for Nuclear Energy Science and Technology (INEST) workshop on Back End Strategies for Dealing with Spent TRISO Fuel, held in Idaho Falls, ID on July 22, 2010.

7.1 Graphite Recycle (ORNL)

Characterization is progressing on the machined, pilot-scale, Graftech recycle PCEA samples (over 200 samples, including “with grain” and “against grain” samples). Properties already determined include density, electrical resistivity, and Young’s Modulus. The coefficient of thermal expansion (25-800°C) measurements are still in-progress since each measurement takes ~24 hours.

The twelve rabbit capsules containing the PCEA bend bars which were successfully inserted into HFIR for cycle 427 are still in HFIR, having completed three cycles of irradiation. Six of the rabbit capsules are scheduled for removal after four cycles (6.4 DPA) which will be completed on August 20th. The remaining six rabbits will stay in HFIR for a further two cycles.

7.3 Pyrochemical Reprocessing - METROX Process Development (ANL)

The components needed to fabricate the redesigned anode compartment within the METROX cell, which provides improved electrical contact between the surrogate fuel materials and anode current lead, arrived from the vendors. Fabrication of the anode compartment was completed and the components were transferred into the glovebox along with new reference electrodes, graphite felt, and cathode rods. METROX process testing with the new apparatus is expected to be conducted in early August.

A source of several grams of plutonium-242 oxide, which is needed to investigate the feasibility of the METROX process, has been identified and the specifics of the transfer are being developed. A presentation on “Pyroprocessing Strategies” was made at the INEST workshop on Back End Strategies for Dealing with Spent TRISO fuel.

Appendix I - Status of FY2010 Milestones – July 31, 2010

(Ordered by Task, then by Date)

Milestone number and description	Level	Responsible Organization	Due Date	Completed or % Complete
2.1.1 – Core design optimization methods - Modeling sequence		TAMU		80*
2.1.2 - Core design optimization methods – Algorithm development		TAMU		25*
2.1.3 - Core design optimization methods – Testing and verification		TAMU		0*
5.0.3 – Determine cost and schedule for installation of a system for thermogravimetric measurements of highly radioactive actinides to verify fundamental models	M3	ORNL	11/23/09	11/10/09
5.0.5 – Assessment of modeling approaches for transport and radiation damage	M3	ORNL	2/18/10	2/17/10
5.0.4 – Complete review of mechanisms for silver transport in the particle and palladium transport and attack on SiC coating	M2	ORNL/UCB	9/30/10	85
5.1.1 – Investigate compound energy formalism model of plutonia with selected transuranics and first order model database	M3	ORNL	5/27/10	5/27/10
5.2.1 – Design a series of benchmark experiments to provide validation & testing data for the presumed mechanisms of Ag transport through pyrolytic C/SiC/PyC layers	M3	ORNL/UCB	8/26/10	98
5.2.2 – Provide model-based preliminary estimate of the impact of fission products on the lifetime of TRISO fuel based on chemical attack by fission products through transport through the kernel and fission product release due to transport mechanisms in SiC	M3	ORNL	8/26/10	90
5.3.1 – Develop physics-informed models for influence of irradiation on TRISO thermal conductivity that is suitable for use in fuel performance codes	M3	ORNL	9/13/10 (Revised)	95
6.1.4 - Initiate facility safety review and modification of procedures and work permits for installation and operation of TRU kernel fabrication equipment in existing glove boxes	M3	ORNL	10/30/09	10/29/09
6.1.5 - Identify candidate sources for heavy metal feedstock needed for TRU kernel fabrication	M2	ORNL	11/20/09	11/20/09
6.1.6 - Complete development of process for fabrication of kernels with internal dispersed oxygen getter	M3	ORNL	3/31/10	3/22/10
6.1.7 - Complete installation of kernel manufacturing equipment in existing glove box	M3	ORNL	7/15/10	7/14/10
6.1.8 - Demonstrate manufacture of Zr/Y surrogate kernels using glove box kernel fabrication facility	M2	ORNL	9/30/10	5
6.1.9 - Acquire heavy metal feedstock needed to start TRU kernel fabrication in FY11	M3	ORNL	9/30/10	80
6.2.5- Initiate facility modification and safety approvals for operation of a fluidized bed coater and associated peripherals using glove box containment for TRU materials	M3	ORNL	11/23/09	11/10/09
6.2.6 - Complete modification of coating equipment for glove box operation	M3	ORNL	7/31/10	7/30/10
6.2.7 - Procure glove boxes for TRU-TRISO coating	M2	ORNL	9/15/10	65
6.2.8 - Complete safety analysis and approval for coating glove box operations	M3	ORNL	9/30/10	Deferred to FY2011
6.4.1 - Design and install an upgraded system for high temperature measurement of creep properties	M3	ORNL	11/23/09	11/20/09
6.4.2 - Complete initial planning for ZrC irradiation effects study	M3	ORNL	6/30/10	6/30/10

6.4.3 - Complete additional ZrC properties measurements	M3	ORNL	9/30/10	65
6.4.4 - Report on extended study of interaction of metallic fission products with ZrC	M3	UNLV	6/30/10	6/30/10
6.4.5 - Complete proton irradiation of ZrC at a temperature of 1400°C up to 2 dpa and subsequent analysis of microstructure	M3	UW-M	6/30/10	66
6.4.6 - Attempt 1500°C for 500 hr Ag/ZrC diffusion couple study		UW-M	6/30/10	6/30/10
7.2.2 – White paper on collaboration opportunities with GIF/Carbowaste	M3	ORNL	12/17/09	12/17/09
7.2.1 – Complete rabbit capsules for insertion in HFIR	M3	ORNL	2/26/10	1/27/10
7.2.3 – Report on completed study on recycled graphite.	M3	ORNL	8/27/10	90
7.3.1 – Interim progress report on METROX studies		ANL	5/21/10	5/21/10
7.3.2 – Report documenting used TRISO fuel treatment process development		ANL	9/30/10	82

* Update information not available.

Appendix II – Project Participants

Deep Burn Task	Name	Organization
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Fuel Cycle Integration	Chris Hamilton	Logos
TRU Fuel Modeling	Ted Besmann	ORNL
Thermo-chemical Modeling	Ted Besmann Marius Stan	ORNL LANL
Actinides and Fission Product Transport	Roger Stoller Paul Schuck John Wills Sven Ruden Giulia de-Lorenzi-Venneri Art Voter Duane Wallace Todd Allen Dane Morgan Izabela Szulfarska	ORNL ORNL LANL LANL LANL LANL LANL Univ. Wisconsin, Madison Univ. Wisconsin, Madison Univ. Wisconsin, Madison
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TRU Fuel Qualification	John Hunn Gary Bell	ORNL ORNL
TRU Kernel Development	Rodney Hunt Valmor de Almeida Jack Collins	ORNL ORNL Subcontract
Coating Development	Jim Miller Jerry McLaughlin Brian Jolly Kevin Cooley	ORNL ORNL ORNL ORNL
Characterization Development and Support	Rodger Martin Steve Nunn Dixie Barker Ivan Dunbar Andrew Kercher Chinthaka Silva	ORNL ORNL ORNL ORNL ORNL Univ. Tennessee, Knoxville Oak Ridge Associated Universities
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