Project No. 09-813

TRISO-Coated Fuel Durability Under Extreme Conditions

Reactor Concepts R&D

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NEUP Nuclear Energy University Programs
U.S. Department of Energy

FINAL PROGRESS REPORT

Project Title: TRISO-Coated Fuel Durability Under Extreme Conditions

Covering Period: October 1, 2009 through September 30, 2013

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Recipient: Colorado School of Mines
MME Department
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Project Objective

The PIs propose to examine TRISO-coated particles (SiC and ZrC coatings) in an integrated two-part study. In the first part, experiments will be performed to assess the reaction kinetics of the carbides under CO-CO_2 environments at temperatures up to 1800ºC. Kinetic model will be applied to describe the degradation. Scanning and transmission electron microscopy will be employed to establish the chemical and microstructure evolution under the imposed environmental conditions. The second part of the proposed work focuses on establishing the role of the high temperature, environmental exposure described above on the mechanical behavior of TRISO-coated. Electron microscopy and other advanced techniques will be subsequently performed to evaluate failure mechanisms. The work is expected to reveal relationships between corrosion reactions, starting material characteristics (polytype of SiC, impurity concentration, flaw distribution), flaw healing behavior, and crack growth.

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Background

Corrosion Behavior TRISO-coated fuel particles can undergo several forms of corrosion and environmental degradation. The fission products palladium and cesium and lanthanides are known to diffuse through the carbon layers and react with the silicon carbide layer leading the formation of localized reaction products, loss of mechanical integrity, and short circuits for fission product release. In addition, reaction between the oxide fuel and carbon buffer can release carbon monoxide that can cause active oxidation of the silicon carbide. Although the kinetics of these reactions are relatively slow, these reactions limit the maximum use temperature and level of burnup that can be achieved with high temperature gas-cooled reactors (HTGRs).

There have been a number of useful studies on the kinetics of reactions between SiC and CO. However, the specific rate-limiting mechanism of the CO-SiC reaction is not known. In the absence of radiation, it appears that the reaction is rate-limited by a surface chemical reaction, possibly by the rate of dissociation of the CO covalent bond. Furthermore, SiC exists as three phases (hexagonal or α, and cubic or β, and amorphous) and hundreds of polytypes that are distinguished by the ordering of stacking faults. Therefore, it is important to recognize that the phase and microstructure can influence reaction rates and degradation mechanisms. For example, stacking faults and grain boundaries can be attacked preferentially. Little is known about the kinetics of reactions of CO with ZrC.

Mechanical Integrity The overall performance of TRISO-coated fuel particles depends on their mechanical integrity, and thus, it is desired to minimize the fraction of particles which experience cracking or crushing. Stresses arise under elevated temperature and severe irradiation, and the failure may further be affected by gaseous corrosive effects. It is well-established that a dominant failure mechanism in TRISO-coated particles begins with irradiation-induced strains in the pyrolitic carbon (PyC). Specifically, it has been shown that shrinkage of the inner pyrolitic carbon (IPyC) due to irradiation may lead to debonding between the IPyC and SiC, and may incur cracks within the IPyC. These cracking mechanisms may lead to a transfer of gas pressure from the IPyC layer to the SiC, thereby increasing the tangential (hoop) stress within the SiC layer. The behavior of SiC is generally elastic until a critical fracture stress is achieved. On the other hand, the IPyC and the outer pyrolitic carbon (OPyC) experience stress relaxation through inelastic creep at elevated temperatures, and these must be taken into account when predicting the overall stress evolution during reactor operation. It is noted that the same failure mechanisms (i.e., IPyC failure) operate when ZrC is used in place of SiC. In addition to elastic and creep deformation of the PyC layers, their elastic response is anisotropic, and this adds a further complexity in developing an accurate thermomechanical model. It has been shown that these effects are multi-dimensional and thus, a one-dimensional model will not provide accurate predictions; such a multi-dimensional model has been developed at Idaho National Laboratory.

Based on the above descriptions, the initial stages of failure of TRISO-coated fuel particles, namely, the thermo-mechanical loading that leads to IPyC failure, is reasonably well-understood. However, the effect of CO/CO_2 and other corrosive species on the progressive degradation of SiC (or ZrC) are not well-understood. The present proposal seeks to develop a better understanding on the coupled effect of irradiation, temperature and corrosion influence degradation in SiC and ZrC.

Status

Task 1: Corrosion Behavior

The work performed to examine the behavior of SiC and ZrC in CO/CO_2 corrosive environments was not published, but is summarized in previous reports. The main outcome is that corrosion may be important in the presence of Cs, but before addressing it, diffusion of Cs must first be understood. Thus, the remaining studies focused on Cs diffusion in SiC. Diffusion and crystallization studies have been performed as detailed in the two journal articles generated.

Papers and Submissions

D. D. Osterberg, J. Youngsman, R. Ubic, I. E. Reimanis, and D. P. Butt "Recrystallization Kinetics of 3C Silicon Carbide Implanted with 400 keV Cesium Ions" *Journal of American Ceramic Society, 96[10] 3290-3295* (2013).

J. Youngsman, B. P. Gorman, I. E. Reimanis, and D.P. Butt, "Diffusion of cesium in ion-implanted β-silicon carbide" submitted to *Journal of Nuclear Materials,* March 2014.

Task 2: Mechanical Characterization

1. Task Status:

Crush tests on SiC hemispheres were performed throughout the project. Though a technique previously developed was employed to interpret the test, it was discovered in the project that this previous test had significant challenges that had not been addressed. Much of the remainder of the project focused on illucidating some of the challenges and developing a framework to interpret crush test results. These results are presented in the MS thesis of Brian Campbell Davis as well as two journal articles which are still in production and should be submitted by the end of April.

Submissions

Brian Campbell Davis, Fracture Strength of the SiC Layer in TRISO Coated Fuel Particles, MS Thesis, Colorado School of Mines May 2013.

B. C. Davis, L. Ward, B. Gorman, D. Butt, B. Fillery, and I. E. Reimanis “Mechanics of a crush test for TRISO particle testing: issues of asphericity”, to be submitted to Journal of Nuclear Materials (2014).

B. C. Davis, L. Ward, D. Butt, B. Fillery, and I. E. Reimanis “Mechanics of a crush test for TRISO particle testing: stress evolution”, to be submitted to Journal of Nuclear Materials (2014).

Milestone Status Table (note: incomplete tasks are ones associated with irradiated particles)

Milestone/Task Description	Planned Completion Date	Actual Complete Date	Percent Complete
Task 1.1	30-Sep-10	NA	100
Corrosion Behavior of Bulk Carbides			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 1.2	31-Oct-11	NA	100
Corrosion Studies of Unirradiated TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 1.3	31-Oct-11	NA	100
Characterization of Unirradiated TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 1.4	30-Sep-12	NA	0
Corrosion Studies of Irradiated TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 1.5	30-Sep-12	NA	0
Characterization of Irradiated TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 2.1	31-Dec-10	NA	100
Nanoindentation and Crush Testing of As-received TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 2.2	31-Dec-11	NA	100
Nanoindentation and Crush Testing of Corroded TRISO Particles			
Milestone/Task Description	Planned Completion Date	Actual Completion Date	Percent Complete
Task 2.3	30-Sep-12	NA	0
Nanoindentation and Crush Testing of Corroded and Irradiated TRISO Particles			

Budget Data

The quarterly approved spending plan budgets were obtained by subtracting from the annual budget the subcontract to BSU for each quarter and dividing that into four equal parts corresponding to each quarter of the year. The first quarter also contains the BSU subcontract amount. The CSM-indirect costs for the BSU subcontract are spread through the four quarters. This spreadsheet is an estimate of actual costs and spending. Please note that this table has been updated since the previous budget estimates and more accurately actual expenditures to date.

	Approved Spending Plan		Actual Spending	
	Quarter	Cumulative	Quarter	Total to Date
To				
12/31/09	200,750	200,750	93.95	93.95
3/31/10	45114	245,864	47,065.76	47,159.71
6/30/10	45113	290,977	75,044.50	122,204.21
9/30/10	45113	336,090	118,839.48	241,043.69
12/31/10	200242	536,332	58,015.95	299,059.64
3/31/11	41427	577,759	34,679.20	333,738.84
6/30/11	41427	619,186	103,338.05	437,076.89
9/30/10	41427	660,613	88,033.58	525,110.47
12/31/11	202841	863,454	5,508.38	530,618.85
3/31/12	43136	906,590	30,086.60	560,611.50
6/30/12	43136	949,726	189,010.06	749,715.51
9/30/12	43136	992,862	18,450.50	768,072.06
12/31/12	202841	992,862	86,995.33	855,161.34
3/31/13	0	0	(355.29)	854,806.05
6/30/13	0	0	14801.97	869,608.02
Totals	992,862	992,862	123,253.98	992,862.00