An Overview of CERMET Fuel Development

...at Marshall Space Flight Center

Marvin W. Barnes
NASA MSFC
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My Background

• Academic
  – BSE in Chemical and Materials Engineering
  – MS in Material Science (in progress)

• Employment
  – United Space Alliance
    • R&D Space Shuttle SRBs Thermal Protection System (TPS)
  – NASA
    • Solid Propulsion Division
      – Launch Abort System
    • Metal Joining and Processes
      – Nuclear Thermal Propulsion
      – Fuel Development
Solid Propulsion

SLS

Orion and Launch Abort System
How Does Nuclear Thermal Propulsion (NTP) Work?

- Propellant heated directly by a nuclear reactor and thermally expanded/accelerated through a nozzle
- Low molecular weight propellant – typically Hydrogen
- Thrust directly related to thermal power of reactor: 100,000 N ≈ 450 MW\text{th} at 900 sec
- Specific Impulse directly related to exhaust temperature: 830 - 1000 sec (2300 - 3100K)
- Specific Impulse improvement over chemical rockets due to lower molecular weight of propellant (exhaust stream of \(O_2/H_2\) engine runs much hotter than NTP)

NERVA Nuclear Thermal Rocket Prototype
Radioisotope

Pu-238 → U-234

Heat Energy = 0.023 MeV/nucleon (0.558 W/g Pu-238)
Natural decay rate (87.7-year half-life)

Long history of use on Apollo and space science missions
44 RTGs and hundreds of RHUs launched by U.S. during past 5 decades
Heat produced from natural alpha (a) particle decay of Plutonium (Pu-238)
Used for both thermal management and electricity production

Fission

Neutron

Fissile Nucleus (U-235)

Heat Energy = 0.851 MeV/nucleon
Controllable reaction rate (variable power levels)

Used terrestrially for over 70 years
Fissioning 1 kg of uranium yields as much energy as burning 2,700,000 kg of coal (>20 GW-hr)
One US space reactor (SNAP-10A) flown (1965)
Former U.S.S.R. flew 33 space reactors
Heat produced from neutron-induced splitting of a nucleus (e.g. U-235)
At steady-state, 1 of the 2 to 3 neutrons released in the reaction causes a subsequent fission in a “chain reaction” process
Heat converted to electricity, or used directly to heat a propellant
Typical First Generation NTP Reactor Design

NERVA Reactor Cross Section

- Control Drums
- Reflector
- Core
- Control Drum Absorber Plate

Fuel Segment Cluster

- Fuel Element
- Support Element
- Inner Tie Tube
- ZrH Moderator
- Outer Tie Tube
- Insulator
- Tie Tube Support
- Collar and Cap
- Miniarch
MSFC Fuels Laboratory Capabilities

- Facilities/Capabilities stand up FY12-13
  - Three new laboratories brought on line with power and exhaust system facility modifications
- All fuel fabrication laboratories are licensed by NRC for handling dU/natU
- All MSFC DU fabrication processes have been approved by the RSO and are operational
- Nuclear Regulatory Commission (NRC) performed a spot inspection of the laboratories with no findings
- MSFC is now equipped to fabricate CERMET fuels from feedstock acceptance through HIP fabrication, testing and characterization
Feedstock Development

- Development of UO$_2$ and surrogate powders focused on particle size, shape, density and stoichiometry
  - 2kg of angular UO$_2$ purchased from Y-12
    - Not optimal for post HIP microstructures
    - Not optimum for CVD W coating process
    - Fine particles, <5μm, clumps and does not flow well

- 3.3kg of spherical UO$_2$ procured from ORNL
  - Qualified Sol-Gel process for TRISO fuels
  - Required development to produce the required size, 100μm
  - Good spherocity and with a tight size distribution

- 3kg of spherical UO$_2$ procured from INL-CSNR
  - Internal gelation being developed by INL-CSNR
  - First UO$_2$ powders produced for NCPS development
  - Very tight size distribution, good shape
Chemical Vapor Deposition (CVD)

- W coated UO$_2$ matrix will increase the life of CERMET fuel
  - Uniform distribution of fuel throughout the matrix
  - Eliminates agglomeration and increases structural integrity of fuel
- No commercial/govt facilities doing W coated UO$_2$ using WCl$_6$
- Currently on Gen 3 of the MSFC CVD system
Net Shape HIP Development

- Design and optimization of full and subscale HIP cans
- Can assembly, fill, closeout, machining and etching
- HIP cycle parameter development and HIP chamber tooling
- Equipment optimization to handle full scale HIP cans
- MSFC HIP system refurb for UO₂ HIP

W-ZrO₂ Post Mo etching and HIP can grinding. Sample ready for CFEET testing (Left)

W-UO₂ sample post HIP

SEM images of W-ZrO₂ cross section

W-UO₂ CFEET sample. Agglomeration of UO₂

HIP can Mo rod stack up prior to assy.

Full scale, 61 channel HIP can failed during cycle due to embrittlement of Nb can material.
Subscale H₂ Test System - CFEET

- A system capable of testing subscale fuel elements at 3000k in flowing H₂ is required for fuel development
- Capable of multiple heating cycles per day for rapid data on fuel integrity
- Requires <100g of UO₂ versus 2.8kg for a full length (17.8” L) element-$$$$
- If the fuel cannot survive CFEET it will not survive NTREES
- Currently operating a 2nd generation CFEET system at MSFC
  - 15kw and 50kw pwr supplies available
  - Obtained 3695K with pure W sample in flowing Ar with 15kw
  - W-ZrO₂ 7 channel sample reached 2338K in a 30sec shakeout test with 50kW
  - Continuing to optimize the system with 50kw and prepping for the W-UO₂ test in Feb ‘14

308 Stainless Steel Samples
Tungsten Rhenium Hafnium Nitride Samples
Tungsten, Graphite (L to R) tested in flowing hydrogen
Above/Left: Pure W sample post shakeout run 2. Sample reached melting point (3695K) and was held in place by the BN insulator. BN insulator had to be destroyed to remove the sample.

View looking down into the CFEET chamber during run 1. BN insulator and bright orange sample inside.

Cut away of CFEET Test Chamber

Upgraded CFEET chamber and 50 kW power supply
NTP CERMET Fuel Fabrication Study

Marvin W. Barnes¹, Dr. Dennis Tucker¹, Lance Hone² and Steven Cook²

¹Metals Engineering Division, NASA Marshall Space Flight Center
²Center for Space Nuclear Research
Presentation Overview

- GE710 Program
- Fuel Compact Fabrication Study
- Tungsten Powder Coating
- Spark Plasma Sintering
- Experimental Approach
- Results
- Conclusions and Future Work
- Other Fuel Development Work
Nuclear Thermal Propulsion (FY16)

• Awarded CIF to investigate CERMET fuel development
• Innovation
  – W Powder Coating
  – Spark Plasma Sintering (SPS)
GE710 Program

• Extensive CERMET fuel development program
  – Over 15 million invested from May 1962 to Sept 1968
  – Operated fuel element fabrication line for “reactor-sized” fuel elements
  – Successfully fabricated 40+ W-60vol%UO₂ fuel elements for qual testing
    • Conducted 10 of thousands of hours of qualification testing

• 710 fabrication approach
  – Press and sinter W-UO₂ compacts
  – Machine cooling channels
  – Stack compacts
  – Weld tubes for cooling
  – Weld external cladding

• Program cancelled
  – Before qual completed

AEC Research and Development “710 High Temperature Gas Reactor Program Summary Report” GEMP-600; Vol I; 1969
Fuel Compact Fabrication Study

• Past efforts focused on consolidating full-length elements
  – Particle segregation/Non-uniformity of fuel particles within W matrix
• Interest in exploring 710 approach
  – Stacking and bonding fuel compacts
• Conducted compact fabrication study
  – Fabricate compacts with high density and uniformly disperse fuel particles
  – Utilizing new process and fabrication technique
    • W powder coating
    • Spark Plasma Sintering (SPS)

Tungsten Powder Coating

- Straightforward approach to particle coating
- Conducted experiments with 6 different organic binders
- Coating Process
  - Blend W powder, dUO$_2$ particles, and binder
  - Stir mixture above binder drop point on hot plate for 5 min
- Improved fuel particle dispersion
  - Coating not as uniform as CVD coated particles

Spark Plasma Sintering

- Simple Process
- Rapid Consolidation/Sintering
- Net-shape/Near Net-Shape Parts
- High Density Parts
- Compatible with W powder coating

1. Pictures courtesy of UC Davis and Substech
Experimental Approach

• Utilized SPS system at CSNR to sinter W/\text{UO}_2 samples
  – Used W powder coated particles
• Sintered total of 24 samples (20 mm diameter; 6 mm thick)
  – Varied peak temperature 1600°C, 1700°C, 1750°C, 1800°C, and 1850°C
  – Held constant 50Mpa axial load with varying
• 20-minute dwell time at peak temperatures
• Measured density and observed microstructure using SEM
Results

- Density
  - Increased with peak sintering temperature
  - Near theoretical density

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**Results**

- **Density**
  - Density can be tailored to meet material performance requirements

![Graph showing the relationship between Max Temp (°C) and % of Theoretical Density](image)

*Max Temp vs % Theoretical Density*

- % of Theoretical Density:
  - 97.0%
  - 98.0%
  - 99.0%
  - 100.0%

- Max Temp (°C):
  - 1600
  - 1650
  - 1700
  - 1750
  - 1800
  - 1850

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*Image credits to NASA*
Results

- SEM
  - Improved microstructure
  - UO$_2$ particles more uniformly dispersed
  - Cross-section depicts some particle elongation

Results

- Energy-dispersive X-ray spectroscopy (EDS)
  - No unexpected phases
Conclusion and Future Work

- Improved density and microstructure
- Further characterization needed and planned
  - Mechanical Properties
    - Hardness Testing
    - Tensile Testing
  - Thermal Properties
  - Analysis
    - TEM/EDS
    - Local Electrode Atom Probe (LEAP)
    - Further SE to quantify dispersion
  - Chemistry
  - CFEET testing planned
Other Fuel Development Work

• FY16 Development Efforts
  – Phase I SBIR – Bonding tungsten CERMET compacts
  – Phase I SBIR - Electrolytic method for tungsten coating
NTP Technical Briefing and Continuation Review
Fuel Fabrication & Testing Milestone
September 26, 2017

Marvin W. Barnes, NASA MSFC
Test Specimen Fabrication Process

Exploring GE710 Process

- Machined or SPS cylindrical wafers (5/8” OD by ½” Long)
- Machined seven 0.110” cooling channels
- Machined cylindrical tantalum HIP enclosure
- Stacked wafers with Mo rods & E-beam welded enclosure
- HIP Bonded at 1800 °C and 30,000 psi
- Chemical etch to remove Mo rods
Specimen Fabrication Accomplishments

- Fabricated 12 pure tungsten wafers
- Fabricated 6 pure tungsten SPS wafers
- Fabricated 15 W/ZrO$_2$ SPS wafers
- Fabricated 3 stacked and HIP bond pure tungsten samples
- Developed process to form cooling channels in pure W wafers
- Identified vendor for W/ZrO$_2$ machining
- Developed process to form cooling channels in W/ZrO$_2$ wafers
- Fabricated stacked and bonded pure tungsten sample with cooling channels
- Conducted W/ZrO$_2$ microscopy and W powder coating optimization
- Conducted W/dUO$_2$ LEAP analysis at INL
• System Modifications
  – Redesigned sample loading apparatus
  – Designed, fabricated and tested tungsten susceptor
  – Optimized induction coil and power supply for operation with susceptor
  – Upgraded insulator and pedestal design
CFEET Test Preparation

• Instrumentation Upgrades
  – Replaced data acquisition unit
  – Installed and verified Williamson pyrometer 650 – 3000 °C (823 – 3273 K)
  – Procured FAR pyrometer
  – Installed Basler networking camera

• Verifying Functionality
  – Developed thermal model of SiC test
  – Verified system operation above 2719 K sample temp (above 2800 K susceptor temp)
  – Mitigated chemical compatibility anomaly
CFEET Test Preparation

- Characterized 2 induction coils
- Conducted 5 pyrometer verification tests
- Conducted 25 steady-state tests (30 minute hold at peak temp)
- Tested various materials
  - Refractory Carbides
    - SiC, ZrC, NbC, TaC, Tricarbides
  - Refractory Metals
    - W, Nb, Hf, Zr
  - Refractory Oxides
    - ZrO$_2$, HfO$_2$
- Conducted 5 W/ZrO$_2$ tests
- Conducted 2 tricarbide tests
- Conducted sintering trials in CFEET
- Optimized transformer ratio and capacitance
- 40+ tests conducted
• Test Date: Aug 28, 2017

• Test Description:
  – Exposed material to simulated environment (elevated temperature and pure hydrogen) in the Compact Fuel Element Environmental Test (CFEET) system using the following parameters:
    • Hold Temperature: 2500K (24% power level)
    • Hold Time: 45 minutes
    • Hydrogen Flow Rate: 5 SLPM
Temperature and Power Profile

Pure Tungsten (W) Test Run in CFEET (45 minutes @ 2500 K)
• Test Date: Aug 30, 2017
• Test Description
  – Exposed material to simulated environment (elevated
temperature and pure hydrogen) in the Compact Fuel Element
Environmental Test (CFEET) system using the following
parameters:
    • Hold Temperature: 2500K (24% power level)
    • Hold Time: 45 minutes
    • Hydrogen Flow Rate: 5 SLPM
• Results for Pure Tungsten Specimen:
  – No macroscopic or microscopic degradation noted
  – No macroscopic or microscopic wafer debonding observed
  – No significant change in mass (less than 0.04%)
  – Increase in luster/sheen of surface due to hydrogen “cleaning” effect

• Future Work for Specimen
  – Scanning Electron Microscopy (SEM)
Temperature and Power Profile

W/ZrO₂ Test Run in CFEET (45 minutes @ 2500 K)
• Results for Tungsten Zirconia:
  – Negligible degradation (some reduction of zirconia may have occurred, further analysis required)
  – No macroscopic wafer debonding observed
  – Minimal mass loss (0.10 g or 0.27%)
  – Increase in luster/sheen of surface due to hydrogen “cleaning” effect
  – No significant microscopy changes

• Future Work for Specimen
  – Scanning Electron Microscopy (SEM)
Conclusions

• Materials and processes show promise, but further development is required
• Additional development needed to assess cladding formation and integrity
• Additional development work to assess bondline integrity
• Additional research needed to develop OD cladding materials
• Further testing need to fully characterize material performance with depleted uranium (microstructure, chemistry, mass loss, etc.)