Study of a Tricarbide Grooved Ring Fuel Element for Nuclear Thermal Propulsion

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Abstract

Deep space exploration, especially that of Mars, is on the horizon as the next big challenge for space exploration. Nuclear propulsion, through which high thrust and efficiency can be achieved, is a promising option for decreasing the cost and logistics of such a mission. Work on nuclear thermal engines goes back to the days of the NERVA program. Currently, nuclear thermal propulsion is under development again in various forms to provide a superior propulsion system for deep space exploration. The authors have been working to develop a concept nuclear thermal engine that uses a grooved ring fuel element as an alternative to the traditional hexagonal rod design. The authors are also studying the use of carbide fuels. The concept was developed in order to increase surface area and heat transfer to the propellant. The use of carbides would also raise the operating temperature of the reactor. It is hoped that this could lead to a higher thrust to weight nuclear thermal engine. This paper describes the modeling of neutronics, heat transfer, and fluid dynamics of this alternative nuclear fuel element geometry. Fabrication experiments of grooved rings from carbide refractory metals are also presented along with material characterization and interactions with a hot hydrogen environment. Results of experiments and associated analysis are discussed. The authors demonstrated success in reaching desired densities with some success in material distribution and reaching a solid solution. Future work is needed to improve distribution of material, minimize oxidation during the milling process, and define a fabrication process that will serve for constructing grooved ring fuel rods for large system tests.

I Foreword

Propulsion systems that derive their power from nuclear reactions have been conceptualized for many decades. Serious efforts to develop this technology were made in both the United States and the Soviet Union. The most well known program was that of the Nuclear Engine for Rocket Vehicle Application (NERVA) that was a joint effort of the U.S. Atomic Energy Commission and NASA. Several engines were built and tested under this program in order to develop this technology for a manned mission to Mars. This work ran until the early 1970’s.¹, ²

The reactors of the NERVA engines consisted of fuel elements formed into hexagonal rods with cylindrical flow passages through which propellant was flowed. The nuclear fission reactions heated the material in the rod and the heat was transferred to the propellant. This type of nuclear propulsion is called nuclear thermal propulsion (NTP). Later, particle bed reactors were considered in a defense project called Timberwind. This reactor geometry flowed propellant through a bed of spherical fuel particles to greatly increase surface area and thus heat transfer. This design showed promise for significantly improving thrust to weight ratios and specific impulse compared to the NERVA engines. It suffered; however, from nuclear thermal instabilities.²

![Figure 1: Illustration of a Grooved Ring Fuel Element](https://ntrs.nasa.gov/search.jsp?R=20180002033 2019-07-29T11:15:49+00:00Z)

Recently, there has been a renewed effort to develop NTP as NASA has been working seriously toward manned Mars mission planned for the 2030s. Much of the fuel fabrication and engine modeling efforts have been focused on the tungsten and graphite based fuel elements in hexagonal rod geometries. Cermet and Graphite based fuels are being studied for fuel element fabrication. This work has been moving toward the hexagonal rod geometry. The authors of this paper have been working on a center innova-
tion fund project to investigate an alternative fuel element geometry proposed by Dr. Bill Emrich at Marshall Space Flight Center, a co-author of this paper. This concept is centered on the idea of increased surface area and heat transfer to the propellant, much as particle bed reactors do, while eliminating the thermal instabilities by creating a defined flow path. This concept is known as the grooved fuel ring element. The idea is to build a fuel element from a stack of washer like rings. Each ring has grooves cut into the surface to allow propellant, constrained by an outer structure, to flow through the grooves to the center where it can flow down and out of the reactor. This provides a large increase in surface area which is directly proportional to heat transfer. These elements would be put together to make up a reactor much in the same way as traditional elements do. Figure 1 illustrates the concept.

In addition to the alternative geometry, the authors are investigating the use of mixed carbide fuels. These fuels are composed of fissile uranium carbide (UC) the fuel and additional refractory metal carbides (e.g. niobium carbide (NbC) and zirconium carbide (ZrC)) in solid solution to increase the melting temperature of UC. The Soviet Union conducted tests of carbide fuels and reached reactor temperature greater than 3000 K. A combination of carbide materials has the potential to allow maximum fuel operating temperatures in the vicinity of 3500 K. This is in contrast to the lower temperature limitations of other fuel forms. The use of carbide fuels could allow the reactor to run at higher temperature and provide more thrust and specific impulse to the propulsion system.

In order to develop this alternate geometry carbide fuel element, data is needed to develop the fabrication process, understand the fuels performance in a hot hydrogen environment and characterize material properties. This has been the primary goal of the work discussed in this paper. Past work has been studied to provide the foundation for this work. Additionally, modeling has been done to understand the neutronics, fluid, and heat transfer processes in such a reactor. Model results have been used to guide material selection and fabrication experiments. Processes and results to date are discussed in this document.

II Modeling of the Neutronics for a Grooved Fuel Ring Reactor

In order to guide the selection of materials for fabrication tests, the reactor concept must be developed. The criticality and operation of the reactor is highly dependent upon material density, material selection, moderating materials, structure, etc. The model was developed with the Monte Carlo N-Particle coede (MCNP). A concept layout was developed for different material combinations for a given power and thrust level. This drove the selection of materials for fabrication experiments and served as a guide for how the manufactured carbides would affect criticality and operating temperature.

II.A Material Selection

The selection of carbides is a trade between high melting temperature properties and low neutron absorption cross sections. While the highest melting temperature are desired, the neutron cross section determines the quantity of uranium (UC) required for the reactor to be critical. The melting point of UC is lower than the refractory carbide compounds of interest; therefore, increasing the UC content lowers reactor temperature limits. Several refractory metals were chosen as the most promising material candidates based on past research.

Figure 2 compares the cross section of Hafnium, Tantalum, Niobium and Zirconium. One can see a significant difference between the materials. While hafnium carbide (HfC) and tantalum carbide (TaC) have the higher melting temperatures, this would be countered by the need for more UC. NbC and ZrC were chosen as the primary candidates of interest due to their lower absorption cross section. Since solid solutions containing ZrC, NbC will capture fewer neutrons, less UC is needed within the reactor. Reducing UC content will ultimately maximize the operating temperature of the fuel by allowing for the highest melting points.

II.B Reactor Model

The reactor model was built using MCNP, a common nuclear physics code. The reactor model employs the grooved ring element design in a cylindrical reactor. Propellant is assumed to be hydrogen. Beryllium is used a moderator in the core and a reflector at the boundary.
The concept reactor was sized for 25,000 N of thrust with a power output of 8 kW/cm$^3$.

Two combinations of carbides were modeled. The first configuration used a fuel mixture of UC, ZrC and NbC. The second configuration changed the mixture to UC, ZrC and TaC. The reactivity of the reactor models were analyzed to determine the amount of fuel required to operate effectively. The first carbide mixture requires a fuel to moderator ratio of 0.261 while the second carbide mixture requires a much higher fuel to moderator ratio of 2.95. Side and top views of these reactor configurations are shown in fig. 3 and fig. 4.

Additionally, calculations were carried out to determine the impact of density on the required quantity of Uranium fuel. The flow paths through the grooves and center of the rings reduce the available volume of fissionable material. Also, the manufacturing process will result in small amounts of porosity. A small amount of porosity is actually required to allow room for the fission products. This is represented in terms of the percent of theoretical density. Theoretical density is 100% fuel in the ring volume. This is plotted in fig. 5. The fraction of the carbide mixture that is UC is plotted against the fraction of theoretical density. One can see that the mixture containing TaC requires much larger amounts of uranium than the NbC mixture. As the density drops below 1, the amount of uranium needed to achieve criticality climbs. Reducing the amount of UC needed is important to raising the operational temperature of the reactor.

While the plot in Figure 5 is done for an enrichment of 95%, a better estimate of enrichment may be determined with further analysis. The less than theoretical density fuel and high neutron cross sections of the other carbides lead toward higher enrichment to counter these effects. Also, less uranium is desired to take advantage of the high melting temperatures of the other carbides. The lower uranium loading as compared with other fuel forms leads to higher enrichment. It may be, however, that if the neutron spectrum is adjusted correctly by varying the amount of moderator in the fuel element, it might be that you can still design to 20% enrichment.

III Thermal/Fluid Model

The thermal and fluid physics in a grooved ring fuel element were modeled in order to better understand the conditions needed for the propulsion system. Comsol was used to create this model. Thermal and fluid physics were coupled together in this model of a representative fuel element. Overall, the authors were looking for the pressure differential driving the flow that would be ideal for heating the propellant as well as determining the number of grooves needed to reach a reasonable mass flow rate. Conditions were adjusted based on the concept engine size of 25,000 N and 8 kW/cm$^3$.

III.A Model Description

The model attempts to represent a fuel element consisting of a stack of grooved fuel rings enclosed by a structure. The stack is limited to two rings in order to reduce computational time. This is expected to be adequate to characterize the physics of the model. The outer structure consists of a beryllium hexagon. The central stack of rings are assigned the material ZrC. As of the writing of this report the required properties of the carbide mixture have not been measured. This will be done in order to update the model at a future date. The fluid is
assigned as H2 and given an inlet temperature of 500 K. Initial temperatures are set near the inlet temperature, except for the fuel which is set to 3000 K. The pressure differential between inlet and outlet is set to 4 psi. The representative picture of the model and the corresponding mesh can be seen in figs. 6 and 7.

III.B Analysis of Results

Several runs of the model were performed to determine the conditions required to drive the flow through the grooves such that the propellant is heated to approximately 3000 K. The pressure differential for this geometry is 4 psi. Ideally the propellant should flow fast enough to remove enough heat that the fuel does not overheat while not moving so fast that the temperature at the outlet is too low. Figure 8 and fig. 9 display the temperature distribution over slices of the element cut horizontally and vertically. One can see in Figure 8 the flow is heated as it passes through the grooves and exits near the max fuel temperature of 3000 K. One can examine fig. 9 to see how the central passage heats up from the flow through the grooved rings. Since this model is limited to two rings, the exit temperature is low near the axis due to the heat sink through the top of the stack to the cold propellant. A more realistic and taller stack would reach a more uniform exit temperature as more flow is heated and a longer path exists for mixing.

The velocity distribution is shown in fig. 10 and fig. 11 as horizontal and vertical slices. Max velocities are on the order of several hundred m/s and the flow remains laminar. There is higher velocity around the corners of the inlet prior to filling the outer passage and passing through the grooves. The flow is seen to accelerate through the grooves and mix in the central passage. As mentioned before a longer more realistic stack will allow for more mixing and uniform conditions at the outlet.

IV Fabrication of a Grooved Ring Fuel Element

The optimal methods for constructing carbide fuel elements is not well understood. Refractory carbides are some of the highest melting temperature compounds making their production very difficult and energy intensive. In previous carbide fuel development programs, produced fuels were very brittle and susceptible to cracking.
It is important to establish the baseline process for the manufacture of solid solution carbides prior to proceeding with fuel element construction. To address this, the authors conducted a series of tests to determine the best processing conditions for manufacture. The authors chose to first employ direct current sintering (DCS) to create sample carbides. Hot isostatic press was determined to be a possible additional step to increase density. After sintering the sample pieces were analyzed to determine density and material distribution. Control variables were adjusted from test to test in order to work toward an optimal density and material distribution.

IV.A Uranium Surrogate and Material Selection

The first step in the processes of learning how to make the carbide fuels was selecting the appropriate materials. As discussed earlier, the neutron cross section has a significant impact upon the engine operation. For this reason, NbC and ZrC were chosen for fabrication experiments. A key component to the carbide fuel is of course the UC. The use of uranium, however, comes with regulatory burdens that translate into additional cost. It was practical to use a surrogate in place of uranium to reduce cost. A surrogate material with a similar crystal structure would behave in a similar manner as uranium carbide in the formation of the material, but offered significant savings and ease of use. Vanadium carbide (VC) was chosen as a surrogate since it is relatively inexpensive and forms a similar crystal structure. The materials tested in the fabrication experiments were NbC, ZrC and VC. The quantities used were approximately 61% ZrC, 31% NbC and 8% VC by weight.

IV.B Powder Preparation

The carbide powders as provided by the vendors were of a nano particle size that varied within some diameter range per quality specifications. Early fabrication experiments in the DCS machine were done with raw powder as provided by the manufacturer. This created some degree of uncertainty in the results since exact particle size distribution is unknown outside of vendor specifications. This occurred due to milling equipment being out of service while being repaired. Some screening of particulate was done in later experiments to reduce the maximum particle size and limit the variability of particle diameter. The micro mill became operational during the summer of 2017. At this point a few samples of powder were milled to reduce particle size prior to the sintering process. The milling process is intended to improve material distribution through the material.

IV.C Direct Current Sintering Experiments

The fabrication of carbide material samples were conducted using a DCS machine. This can be seen in fig. 12. Powder is loaded into a die prior to being placed in the machine. Once installed, the DCS places the sample under a pressurized inert atmosphere. It then runs a large current through the powder to raise its temperature. The machine can be controlled in several ways. Pressure, rise time, dwell time (at max temperature), and cooling rate can all be controlled. These variables were adjusted in an effort to work toward a sample with high density and good material distribution.

IV.C.1 Density of Samples

High densities were achieved earlier than anticipated. The authors were successfully able to achieve densities up to 98%. This is above the required density which is expected to be approximately 95% to accommodate fission product production. Dwell time, cooling rate, and sintering temperature were critical variables to achieving density goals. Sintering temperatures of approximately 1600 °C were adequate to reach high densities (>95%TD). The carbides also achieved high densities at fast cooling rates. In fact the machine was turned off for a maximum cooling rate of approximately 200 degC/min. The highest densities were achieved at this cooling rate. Additionally
dwell times on the order of 20 minutes were adequate to give the best results. Plots of these variables can be seen in fig. 13, fig. 14, and fig. 15. These figures plot the percent of theoretical density as a function of sintering temperature, cooling rate, and dwell time respectively. Note that there is a good degree of variability believed to be in large part due to the variation in particle size.

**IV.C.2 Material Distribution**

In order to achieve carbide fuel elements with uniform properties and best performance the distribution of the carbide elements must be as close as possible to uniform. Particle distribution of test samples were studied in addition to sample densities. Measurements were made using a scanning electron microscope and x-ray spectroscopy. The scanning electron microscope allows one to see the grain structures formed in the carbide sample.

X-ray spectroscopy is employed to identify the materials in the samples and their distribution throughout the crystal structure.

Early samples showed large grain boundaries. In these early samples the particle distribution is less than ideal. Material can be seen to be clustering together. This is believed to be in large part due to the variability in particle size with a non-optimal direct current sintering process contributing. The x-ray spectroscopy at one location of the first sample fun at 1600 degrees Celsius for 10 minutes with a 100 C/min cooling rate can be seen in fig. 16 with table 1 showing the amount of material at the various locations.

After analyzing several samples the authors decided to sift the materials through a screen in order to limit the maximum particle size. This limited the particle size to 45 micron. Material screening showed improvement to the particle distribution. This can be seen in fig. 17 and table 2.

**IV.D Cutting Grooves**

Important to the manufacturing process is the method for creating the grooves in the carbide rings as well as the center hole. Two tools are being investigated for use in this effort; a diamond wire saw and a water jet. The saw was tested on a carbide sample and successfully cut several grooves through the material. The saw has several limitations however. The saw is limits the groove
diameter to the width of the blade. It is difficult to work with due to the small size of the carbide rings. Also, it is not effective at cutting the center hole. The water jet is thought to be a promising candidate. Current water jet capabilities at Marshall Space Flight Center allow the groove width to be cut as small as 0.03 inches. The water jet is capable of cutting curves and can cut out the center hole. Adjusting the pressure should allow the operator to cut the grooves without cutting all the way through the ring. Water jet tests are planned future work. Test cuts will be performed on a carbide sample. This will be followed by an attempt to produce several full size grooved fuel rings with the DCS and water jet.

V Carbide Material Characterization

Characterizing the material properties of the carbides used in this study is necessary to understand their performance in a reactor environment. We have reviewed previous work after reviewing available literature and have built upon past work. The authors are working to measure the thermal diffusivity of the carbide mixtures. Also, the material samples created using DCS are being tested in a hot hydrogen environment (T > 2000K). Testing in a hot hydrogen environment is used to verify the structural integrity and chemical stability of the materials at relevant temperatures while exposed to the proposed hydrogen propellant.

V.A Thermal Diffusivity

Thermal diffusivity measurements were attempted. Two samples were measured by heating the material with a light source to a set temperature. The material is then allowed to cool back to room temperature. The cooling rate is measured and used to determine thermal diffusivity. Multiple measurements were taken and average was found. Thermal diffusivity numbers were obtained and plotted for the two samples. For reasons unknown the samples disintegrated at relatively low temperatures. Testing in a hot hydrogen environment showed survivability at much higher temperatures, so this result is as of yet unexplained. It could possibly be the result of a preexisting fracture or other outside factor. The data obtained is presented in fig. 18. Future work is planned to take new measurements of thermal diffusivity at much higher temperatures.

V.B Hot Hydrogen Environment Testing

Hot hydrogen environmental testing was conducted in the Compact Fuel Element Environmental Test (CFEET) system at NASA Marshall Space Flight Center (fig. 19). CFEET has a 50 kW induction power supply and two-color pyrometers for temperature measurements up to
Figure 18: Thermal Diffusivity Measurements of Two Carbide Samples

3000 °C. It is designed to flow hydrogen across subscale fuel materials for testing at high temperatures for up to ten hours.

Figure 19: Compact Fuel Element Environmental Test system

An initial sample was run at a relatively low temperature to verify if the sample would fall apart similar to observed in the thermal diffusivity testing. The sample maintained structural integrity when exposed to a maximum temperature of 2000 K for 30 minutes. Subsequently, three samples were run for 30 minutes (rough timescale of a single engine burn on a Mars mission) at 2250 K. X-ray diffraction (XRD) analysis appears to show the tricarbides moving toward a solid solution. Solid solutions refer to an ideal mixture of the tricarbides as opposed to discreet clusters of compacted binary carbide powders. This is indicated by the shifting toward a single defined peak at an intermediate 2θ values. This XRD data can be seen in fig. 20. There does however, exist the presence of unidentified peaks post-CFEET testing. Further analysis is needed to verify if unidentified peaks are due to the formation of free carbon, Nb2C, or other lower melting temperature compounds.

Figure 20: X-Ray Spectroscopy Analysis Before and After Testing in CFEET

Additional CFEET tests were conducted once the micro mill was operational. This allowed for carbide samples to be made from milled powders in an effort to produce improved distribution in the sintered carbide pieces. It was observed that cracking was evident in the sintered carbides for powders that had been milled.

Two tests were run on milled carbide units. Each test was run for 45 minutes. The peak operational temperatures were 2500 K for the first test and 2750 K for the second. The formation of blisters as seen in fig. 21 were observed in the samples following exposure to the hot hydrogen environment. The cracks were also seen to worsen(fig. 22).

EDX analysis revealed large quantities of carbon in the blisters. The samples were also shown to contain large amounts of Zirconium Oxide (ZrO2). The formation of oxides in the carbides is likely the cause of the formation of these two features. One can see the XRD analysis, shown in fig. 23 of the tested units, that spikes corre-
Figure 22: Crack formation in carbide samples following CFEET Test

Figure 23: X-Ray Spectroscopy Analysis Before and After Testing in CFEET

Figure 24: XRD Analysis Prior to Milling Carbide Powder

Figure 25: XRD Analysis following Carbide Powder Milling

Zirconium oxide formation hinders the construction of a structurally sound high melting temperature carbide fuel element. For this reason the authors are interested in determining the root cause of the oxide formation and modifying the fabrication process to prevent its formation.

The powders were studied before and after the milling process. It was thought this was a likely cause of the oxide formation since little oxide was seen in the pieces that were not milled. XRD analysis was performed on powders before and after milling. The results of the before and after milling XRD analysis are plotted in fig. 24 and fig. 25. The key material response points are plotted to illustrate materials present.

One can see in fig. 24 that there is no oxide formation. The points indicating ZrO2 are not present. In contrast to this fig. 25 indicates the formation of ZrO2 in the milled carbide powder. This indicates the milling process results in freeing carbon and the formation of ZrO2.

VI Conclusions

This project set out to begin development of the fabrication process of carbide fuels for a grooved ring fuel element of a nuclear thermal rocket. The team ran the initial calculations and models to drive the carbide fuels experiments. The models built will serve as a foundation upon which more thorough modeling can be completed following additional data collection.

Direct current sintering experiments were shown to be quite effective at achieve high density carbide samples and reasonable material distribution. Testing of sintered carbides in a hot hydrogen environment also showed positive results. The tests showed the carbide samples moving toward a solid solution which is optimal for fuel design. They also showed the carbide could withstand the hot hydrogen environment when there was low oxide formation. Although unmilled sintered tricarbide samples performed well when exposed to hydrogen at high temperatures, oxidation of the carbide powders during the milling process resulted in severe cracking and blister formation in the hot hydrogen environment.

The next step in developing the fabrication process is to address the oxidation of the carbide powders during the milling process. The milling process is employed to improve material distribution throughout the sintered carbide mixture by reducing particle size. Oxidation must be prevented in order to develop carbide fuels that will maintain structural and chemical integrity in a hot hydrogen environment as will be seen in a nuclear thermal rocket.

Additionally, experiments need to be run to determine a method for moving the carbide mixtures toward a solid
solution. Heating in a graphite furnace under vacuum may achieve this and is planned for testing. Machining of the grooves and central passage to form the carbides into grooved rings for the construction of a full size element must also be addressed. Tests are planned for cutting the sintered carbides in a water jet. Finally, a full size prototype grooved ring fuel element needs to be built and tested in the Nuclear Thermal Rocket Element Environment Simulator (NTREES) for performance assessment.

The team believes the results of the work conducted in FY17 to be encouraging. The carbide fuels and the grooved ring fuel element are a promising fuel form and design geometry for future nuclear thermal rockets.

A Nomenclature

- $K = $ Kelvin
- $NbC = $ Niobium Carbide
- $ZrC = $ Zirconium Carbide
- $ZrO2 = $ Zirconium Oxide
- $TaC = $ Tantalum Carbide
- $HfC = $ Hafnium Carbide
- $UC = $ Uranium Carbide
- $VC = $ Vanadium Carbide
- $DCS = $ Direct Current Sintering
- $H2 = $ Diatomic Hydrogen
- $XRD = $ X-Ray Diffraction
- $EDX = $ Energy Dispersive X-Ray Spectroscopy
- $NTREES = $ Nuclear Thermal Rocket Element Environment Simulator

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