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Nuclear Thermal Propulsion Reactor Materials

Douglas Burns and Stephen Johnson

Abstract

Nuclear thermal propulsion (NTP) systems have been studied in both the USA and the former Soviet Union since the 1950s for use in space science and exploration missions. NTP uses nuclear fission to heat hydrogen to very high temperatures in a short amount of time so that the hydrogen can provide thrust as it accelerates through an engine nozzle. Benefits of NTP systems compared to conventional chemical and solar electric powered propulsion systems include higher fuel efficiency, greater mission range, shorter transit times, and a greater ability to abort missions and return to Earth in the event of system failure. As a result of these benefits, the US National Aeronautics and Space Administration (NASA) is evaluating NTP for use in crewed missions to Mars, and plans for a possible mid-2020s flight demonstration of a NTP engine are under development. The extremely harsh conditions that NTP systems must operate in present a number of significant engine design and operational challenges. The objective of this chapter will be to describe the history of NTP material development, describe current NTP material fabrication and design practices, and discuss possible future advances in space propulsion material technologies.

Keywords: space, nuclear power, nuclear fuel, high-temperature materials

1. Introduction

The dream of one day expanding humanity’s presence into the solar system will require advanced propulsion systems that provide high levels of thrust and efficient use of fuels. Thrust will be needed to leave Earth’s gravitation field and to establish stable orbits when approaching other planets and returning home. Many of the missions that will one day be of interest to human explorers will require travel to locations that are far away from the sun, so dependence on solar power will not be an option, and prepositioning enough chemical propellant to allow freedom of movement and the ability to return to Earth will be too expensive.

A wide range of studies, including the National Aeronautics and Space Administration’s (NASA) recent Design Reference Architecture (DRA) 5.0 Study [1], have shown that nuclear power can enable exploration of the solar system. Nuclear thermal propulsion (NTP) and nuclear electric propulsion (NEP) are technologies that can provide the necessary thrust and power densities to enter and leave gravity wells of planets, moons, and large asteroids, and they do not need external sources of power to generate propulsion. Heat produced through fission is all that is needed to add energy to a propellant and produce thrust.
Space nuclear reactors rely on nuclear fuels that include a range of fissionable compounds. Uranium oxide (UO$_2$), uranium nitride (UN), uranium carbide (UC and UC$_2$), and uranium oxycarbide (UCO) are ceramic materials that have been studied by various space reactor technology development activities. Each of these materials has advantages and disadvantages related to use in space reactors, but they are all capable of achieving the extremely high temperatures that will be needed to move humans and equipment from Earth to other parts of the solar system.

1.1 Fundamentals of rocket propulsion

The function of a rocket engine is to provide a force $F$ over a time $t$ to a body of mass $m$ in order to change velocity $\vec{v}$ of the body by an amount $\Delta \vec{v}$. The rocket expends a mass $\Delta m$ of fuel in order to complete a velocity change maneuver. The force on the engine is produced by heating a propellant and expelling it through an expansion nozzle at a velocity $v_e$ with respect to the engine. The force produced is given by $F = \frac{\Delta m}{\Delta t} v_e$, where $\frac{\Delta m}{\Delta t}$ is the propellant mass flow rate.

The efficiency of an engine is determined by the force produced by a unit of mass flow rate, which is frequently defined in terms of "specific impulse." Specific impulse is given by $I_{sp} = \frac{v_e}{g}$, where $g$ is the acceleration of gravity (note that $I_{sp}$ has units given by velocity ÷ acceleration = seconds). During a maneuver, the initial mass of the engine $m_o$ changes to a final value of $m$ in order to produce a change in velocity $\Delta \vec{v}$, so that the mass ratio $\frac{m}{m_o}$ is a measure of maneuver efficiency. In free space, with no other forces acting on the engine, conservation of momentum leads to the “rocket equation” given by:

$$ \frac{m}{m_o} = e^{\frac{\Delta \vec{v}}{g}} = e^{\frac{\Delta v_e}{g}} $$

(1)

This equation illustrates how $I_{sp}$ is tied to engine efficiency.

Another important aspect of rocket engine operations is that propellant exhaust velocity $v_e$ is given by:

$$ v_e^2 = \frac{k \sqrt{R T_c} [1 - \left(\frac{p_e}{p_c}\right)^{\frac{k-1}{k}}]}{(k-1)} $$

(2)

where $k$ is a constant given by the ratio of propellant liquid and vapor phase specific heats, $R$ is the universal gas constant, $M$ is the propellant molecular weight, $T_c$ is the combustion chamber temperature, $p_e$ is the nozzle exit pressure; and $p_c$ is the combustion chamber pressure.

As a result, $v_e^2 \propto \sqrt[18]{T_c}$, and therefore $I_{sp} \propto \sqrt{T_c}$, so that engine efficiency increases in systems that produce high temperatures and use low molecular weight propellants. In chemical rockets, the highest available $I_{sp}$ is produced by burning H$_2$ and O$_2$ to produce H$_2$O with a molecular weight of approximately 18 g/mol. Nuclear rockets, on the other hand, use H$_2$ as a propellant, so they produce specific impulses that are approximately $\sqrt{18} = 3$ times higher than the impulses produced by chemical rockets, for a given chamber temperature. Figure 1 shows a comparison of theoretical specific impulses and mass ratios (i.e., ratio of take-off mass to final mass for Earth escape) for various propulsion systems [2].
Nuclear thermal propulsion systems can use a range of fluids for thrust and reactor cooling. Examples include hydrogen, ammonia, methane, octane, carbon dioxide, water, and nitrogen [3]. Specific impulse is lower for higher molecular weight fluids, but the heavier fluids require less storage capacity, and they could be mined, or synthesized, on interplanetary trips.

Nuclear engine design requires iterative consideration of reactor neutron thermal hydraulic and structural characteristics combined with engine system-level performance analysis [4]. Effective design and analysis sequences involve establishing a preliminary core design that meets the fundamental neutronic performance requirements of start-up criticality and reactor control. Fuel element designs using fixed fuel compositions and uranium enrichments are developed early in the design process, and then the preliminary design is used to determine neutron and gamma energy deposition characteristics that feed an integrated thermal hydraulic/structural analysis of the core’s internal components. Once acceptable neutronic and thermal/structural performance is achieved, overall engine performance is evaluated to determine how well the design satisfies mission requirements. The analysis sequence is then revised as necessary to optimize engine performance characteristics to support specific mission profiles.

Engine performance can be improved by various methods of controlling propellant flow through the reactor core and varying fuel compositions. For example, enrichment zoning within the fuel elements, with lower enrichments in high-power regions of the core, can be effective at flattening reactor power profiles and producing more uniform propellant exit temperatures. The cost of these design complications is often slightly reduced core reactivity that can have an impact on engine performance (i.e., specific impulse), but compensation for the reactivity loss is

Figure 1.
Comparison of rocket propulsion system characteristics.
often possible through careful consideration of performance enhancements outside of the reactor fuel (e.g., propellant orificing, reductions in reactor mass, and the use of materials with low neutron absorption characteristics).

A wide variety of fast spectrum and thermal spectrum reactor designs have been developed for use in space propulsion systems. Fast spectrum reactors rely on high-energy (i.e., “fast”) neutrons having average energies greater than 0.5 MeV to produce heat using materials that can fission after fast neutron absorption, while thermal spectrum reactors require the use of moderator materials to slow neutrons down to lower energies that are more readily absorbed. Fast reactors require fuel that is relatively rich in fissile material, while thermal reactors can operate with low-enriched uranium fuels.

Both fast and thermal spectrum reactors are typically designed with reflectors made from materials such as beryllium that prevent neutron leakage from the reactor core without producing a significant amount of neutron absorption. In space reactors, axial reflectors are often placed above and below the reactor core and radial reflectors are often placed around the core to reflect neutrons that would otherwise escape from the core back into the reactor’s fuel. Control drums that are rotated to add enough reactivity to start up the reactor and make minor adjustments to its power profile are typically placed inside the radial reflector. A material with a high neutron absorption cross section (e.g., boron carbide, B$_4$C) is placed on one side of the control drums to remove neutrons while the reactor is shut down. The drums are rotated to move the neutron absorption material farther away from the core in order to start up the reactor.

Fuel depletion and fission product buildup during reactor operation are typically areas of concern for reactor design, but space reactor operating times are typically very short, so fuel burnup and fission product buildup are usually of little importance to NTP reactor designs.

2. Space reactor research and development programs

2.1 Rover/NERVA

Nuclear thermal propulsion systems were studied extensively during the 1950s and 1960s, but they were considered to be too heavy and expensive for deployment. At the time, chemical rockets and solar power were more economical for near-Earth operations that were the focus of the world’s space agencies. However, recent interest in deep space exploration, and especially interest in sending astronauts to Mars, has reinvigorated NTP research for several reasons. First, the longer thrust duration than chemical rockets that can be produced with an NTP system could cut the travel time to Mars by 20–25%. The reduced travel time is important because it would reduce the amount of dose that astronauts would receive from cosmic radiation during the voyage. Second, the higher thrust would extend the available launch window for missions to Mars. Conventional chemical rocket engines can only be used to reach Mars during a 30-day window that opens every 26 months due to the relative positions of the planets, while nuclear propulsion systems can provide enough thrust to leave Earth’s orbit and reach Mars during more points during the orbital profiles of the two planets. Finally, NTP systems could extend the amount of time during which a mission to Mars could be aborted and still allow astronauts to safely return to Earth. Chemical rocket fuel needed for the return trip from Mars to Earth would likely need to be sent to Mars in advance of a crewed mission, so astronauts would have to reach Mars in order to
return to Earth after their initial supply of fuel is consumed, if chemical rockets were used for the mission. An NTP system, on the other hand, would be able to turn around and return to Earth before reaching Mars, if the mission had to be aborted.

Several successful research and development programs focused on space reactor fission power technologies have been established over the past 60 years. The earliest, and most extensive of these efforts, were the Rover and Nuclear Engine for Rocket Vehicle Application (NERVA) programs that were sponsored by the US Atomic Energy Commission (AEC) between 1958 and 1971 [5]. A total of 13 research reactors and 6 nuclear engines were built and tested under the Rover/NERVA programs at the AEC’s Nevada Test Site (NTS) Nuclear Rocket Development Station (NRDS) and other facilities located across the country [6]. The Rover reactor development and testing efforts were led by the Los Alamos Scientific Laboratory (LASL), and the NERVA reactors were designed and built by Westinghouse Electric Corporation (Astronuclear) and Aerojet-General Corporation following a 1961 design competition. The Kiwi (1955–1964), Phoebus (1964–1969), and Peewee (1969–1972) series of reactors were developed and tested under Rover to demonstrate the basics of nuclear rocket technology and to study characteristics of high-temperature nuclear fuels and long-life fuel elements. The NERVA NRX and XE engines were also built between 1964 and 1969 and tested at NRDS to study the complexities of nuclear engine start-up, full-power operation, and shutdown. A list of the best performance parameters achieved during the Rover/NERVA programs is presented in Table 1 [7].

Early work in the Rover/NERVA program was performed with uranium carbide (UC or UC$_2$) and uranium dioxide (UO$_2$) fuel particles embedded into graphite. The effects of fission product interaction with the graphite matrix quickly led to the use of pyrolytic graphite-coated UO$_2$ particles in the Rover/NERVA fuel designs. The pyrolytic carbon contained fission products produced during reactor operations before the fission products could cause dislocations in the graphite structural materials. The pyrolytic carbon layer also protected the UO$_2$ particles from oxidation during fabrication and handling of the fuel.

A major drawback associated with using graphite in space reactors is that graphite converts to methane (CH$_4$) under hydrogen exposure, and the conversion causes the graphite to corrode. During the Rover/NERVA programs, the graphite matrix used in the reactor fuel was protected from corrosion through application of a high-temperature niobium carbide (NbC) coating. However, the use of the NbC coating leads to issues associated with “mid-band corrosion,” which was higher corrosion rates in the center third of the reactor fuel elements where power density

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reactor (test date)</th>
<th>Peak performance achieved</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power</td>
<td>PHOEBUS 2A (July 1968)</td>
<td>4100 MWt</td>
</tr>
<tr>
<td>Peak fuel temperature</td>
<td>PEEWEE (November 1968)</td>
<td>2750 K</td>
</tr>
<tr>
<td>Specific impulse</td>
<td>PEEWEE (November 1968)</td>
<td>848 s</td>
</tr>
<tr>
<td>Maximum restarts</td>
<td>XE’ (June 1969)</td>
<td>28</td>
</tr>
<tr>
<td>Accumulated time at full power</td>
<td>NF-1 (June-July 1972)</td>
<td>109 min</td>
</tr>
<tr>
<td>Continuous operation</td>
<td>NRX-A6 (December 1967)</td>
<td>62 min</td>
</tr>
</tbody>
</table>

Table 1. Maximum performance results achieved during the Rover/NERVA programs.
was high. The corrosion was found to be caused by cracking of the NbC coatings used in the fuel’s hydrogen coolant channels due to a mismatch between the thermal expansion coefficient of the NbC coating and the thermal expansion coefficient of the fuel matrix. The NbC coatings were eventually replaced with zirconium carbide (ZrC) coatings because of zirconium carbide’s superior resistance to fission product diffusion at high temperatures.

The search for solutions to the corrosion problem also led to the development of \{(\text{U, Zr})C, \text{graphite}\} composite fuel. The coefficient of thermal expansion for the composite fuel was 6–6.5 μm/mK (versus 3 μm/mK for fuel made from UO$_2$ dispersed in graphite), which matched the NbC coefficient of 7.1 μm/mK fairly well [7].

Carbide material systems have several favorable features applicable to NTP reactors including:

- Relatively small neutron absorption cross sections
- High melting points
- Thermal stability
- Low volatility
- High fuel densities
- High moderation ratios
- Low material densities (<10 g/cm$^3$)

However, the complex fuel designs used in the early Rover/NERVA tests were challenged by the mechanical loads, thermal stresses, and high radiation fields found in NTP reactors. The intense operational conditions contributed to the formation of stress fractures in the fuel coatings and surfaces and the cracking encouraged increased hydrogen penetration into the fuel that produced fuel degradation.

The Small Nuclear Rocket Engine (SNRE) was the last engine design studied by Los Alamos National Laboratory (LANL) under the Rover/NERVA program. The SNRE was a nominal 16,000 lbf thrust engine that was originally intended for short-run-time, unmanned missions, and the SNRE stage design was constrained to fit within the payload volume of the planned space shuttle. The reactor’s core design used hexagonal fuel elements and hexagonal structural support elements (i.e., tie tubes), and the number of elements could be varied to support different thrust requirements. Higher thrust designs for SNRE meet or exceed performance characteristics identified in the DRA 5.0 study, so SNRE would be suitable for use in human missions to Mars.

2.2 GE-710 high-temperature gas reactor research and development program

The GE-710 [8] high-temperature gas reactor (HTGR) and the Argonne National Laboratory (ANL) nuclear rocket engine programs [9] focused on development of ceramic-metal (cermet) fuels consisting of uranium ceramic material (e.g., uranium dioxide [UO$_2$] or uranium nitride [UN]) embedded in a refractory
metal matrix (e.g., tungsten). To ensure good bonding between the kernels and the matrix, the kernels were coated with a thin layer of the matrix metal (i.e., tungsten [W] or molybdenum [Mo]). In addition, the coolant flow channels of the cermet fuel were coated with either tungsten or niobium.

The GE-710 program ran from 1962 to 1968 with the objective of performing reactor tests of a closed-loop system (i.e., an engine system that recycled engine propellant) that used neon as a coolant, and an open-loop system (i.e., an engine system that expelled the reactor coolant to produce thrust) that used hydrogen as the reactor coolant. Final program goals focused on longer-term operation (approximately 10,000 h) at fuel temperatures in the 2000–2250 K range. Major achievements during the GE 710 program included down selection to either W-UO$_2$ or Mo-UO$_2$ cermet fuels, significant development of fabrication and brazing techniques for cermet fuel elements, development of sintering methods for fabrication of high-density fueled cermets, and initiation of in-pile testing. Molybdenum was also investigated as a substitute for the tungsten matrix, but the lower strength of Mo caused increased fuel swelling at high burnups due to fission gas buildup. The loss of Mo due to vaporization at high temperatures during electron beam welding and during thermal cycling was also undesirable.

The W-UO$_2$ cermets tested under the GE-710 HTGR program were cold pressed and sintered into segments of approximately 12.7 mm lengths. Tungsten powder composed of 1–2 μm diameter particles with a uniform spherical shape was used in the sintering process. These particles produced increased sintered densities (e.g., 95% of theoretical density) compared to coarser particles with predominantly angular or planar shapes. Microspheres are a desirable particle shape because they have good heat transfer characteristics, they have consistent grain sizes, they are non-abrading (and therefore dust free), they are free-flowing, and they can be engineered to be either soft or hard. Conversely, powders with varying grain sizes are undesirable because they tend to agglomerate, they can be abrasive, and they have low reproducibility.

After sintering, the GE-710 fuel segments were machined into a hexagonal shape. Coolant channels with 0.914 mm (0.036 in) diameters were drilled into the segments, and then 0.203 mm (0.008 in) wall thickness coolant tubes were sealed into place on one end of the segments by tungsten inert gas (TIG) welding. A header was brazed to the other end prior to complete element assembly, and a tantalum (Ta) spacer plate was used adjacent to the header to protect the fuel from the braze material. The segments were placed into a 0.381 mm (0.015 in) wall thickness cladding, and the fuel was bonded to the cladding using a high-temperature, high-pressure autoclaving process. Autoclaving was typically carried out at a pressure of 10.3 megapascals (MPa) (1494 pounds per square inch [psi]) and 1922 K for 1 h, although an alternate hot-gas pressure process was also used at 68.9 MPa (9993 psi) and 2022 K for 2–3 h.

Dissociation of UO$_2$ into free uranium and hyperstoichiometric UO$_2$ or oxygen during the sintering process had a detrimental effect on fuel fabrication. Fuel performance issues arose from UO$_2$ dissociation because an increase in excess oxygen within the fuel led to an increase in fuel swelling, and the excess oxygen could react with the W matrix to form WO$_2$ stringers in the matrix grain boundaries. Free uranium was also detrimental to fuel dimensional stability and caused negative reactions with the cladding materials that were used. There was little to no mobility of free uranium below the fuel particle melting temperature range (1422–1644 K), but uranium formed a two-phase mixture that produced fuel swelling above the melting temperature range.
Challenges associated with dissociation of UO$_2$ were first addressed by the addition of thorium oxide (ThO$_2$) as a stabilizing compound, but testing showed that ThO$_2$ only delayed free uranium migration. A more suitable solution to UO$_2$ dissociation was found to be the addition of substoichiometric UO$_2$ combined with the ThO$_2$ stabilizer since substoichiometric UO$_2$ retained a single phase as temperatures increased. Dissociation and free uranium migration became an issue only during thermal cycling, and oxygen to uranium ratios of 1.984–1.988 were found to have the best performance during thermal cycling tests [10].

Differences in the coefficient of thermal expansion between the fuel, matrix, and cladding also presented themselves during the development program. During bonding of the cladding to the fuel, the cladding material expanded two times more than the matrix material, resulting in compression at the interface when the fuel was cooled. Alloying with 3 wt% rhenium (Re) in the tungsten matrix increased the low-temperature ductility of the matrix.

The most promising clad materials used in the GE-710 program were elemental tantalum, tantalum alloys (T-111 and tantalum-10 weight percent tungsten [Ta-10W]), and a tungsten-30 weight percent rhenium-30 weight percent molybdenum (W-30Re-30Mo) alloy. Tantalum was selected as the initial cladding material because the material was readily available, and it had sufficient compatibility with the W-UO$_2$ cermet fuel. However, tantalum clad performance was limited by free uranium that formed reaction voids in the cladding. The voids formed because repeated cycling of the fuel allowed uranium metal to precipitate out of single-phase UO$_2$. The uranium migrated through the W matrix grain boundaries and into the Ta cladding, and leak paths developed as the uranium metal re-oxidized to form UO$_2$. The T-111 cladding material was attractive because the alloy maintains a fine-grained structure that limits uranium movement until grain growth occurs above 1922 K. However, the alloy has a high oxygen permeability that results in reaction void formation. The W-30Re-30Mo alloy used in the later stages of the GE-710 program was found to have low oxygen permeability, low sensitivity to gas impurity absorption, high strength, high melting point, and good ductility. Unfortunately, high bond stresses caused by thermal expansion mismatch between the W-30Re-30Mo clad and the fuel matrix occurred during thermal cycling. An anneal heat treatment was used to overcome the bond stresses, but the treatment caused re-precipitation of the sigma phase at grain boundaries which led to clad weakness. Volatilization of Mo at high temperatures also increased sigma phase formation and reduced clad strength.

Irradiation tests performed under the GE-710 program included tests of UO$_2$ and ThO$_2$-stabilized UO$_2$ fuel samples in the Idaho National Laboratory’s (INL) Engineering Test Reactor (ETR). Matrix materials used in the samples included W, W-Re, and Mo with Ta-10W, W-30Re-30Mo, and niobium (Nb) cladding. Approximately half of the samples evaluated in the ETR testing campaign developed fission gas leakage. Further testing was performed in the Low-Intensity Test Reactor (LITR) at the Oak Ridge National Laboratory (ORNL) using W and W-Re matrix material with W-30Re-30Mo and W-25Re-3Mo cladding. The results of the test were similar to the ETR results. However, a third series of tests in the Oak Ridge Research (ORR) reactor with basically the same matrix-cladding combinations showed significant improvements that were achieved by reducing the density of UO$_2$ in the fuel to provide void space for fission product gas accumulation.

Fuel failure modes observed during the GE-710 testing included [7]:

- Loss of oxygen from UO$_2$ at high temperatures followed by the formation of substoichiometric UO$_2$, free uranium, and uranium penetration of the cladding wall during thermal cycling.
• Volume expansion, and eventual cracking of the W-UO$_2$ fuel matrix, during very high-temperature operation after significant thermal cycling.

• Void formation between the cermet fuel and the fuel cladding during fabrication and early operation.

• Fission product damage/release after 4000–7500 h of operation at 1870–2270 K in fuel specimens sintered to 95% or greater theoretical densities.

• Preferential vaporization of Mo and other lower melt point materials out of the clad at temperatures above 2470 K. Molybdenum was found to be a poor candidate for alloying because Mo vapor pressure becomes significant at temperatures above 2470 K.

Physical mechanisms determined to have caused the failure mechanisms included:

• Transparency of Ta and Ta alloy materials to oxygen at intermediate and high temperatures.

• Volume expansion and cracking caused by incompatibility of coefficients of thermal expansion between tungsten and UO$_2$ in the fuel matrix. The incompatibility caused fuel particles to pull apart from the tungsten matrix at high temperatures and after multiple thermal cycles.

• Void formations caused by difficulties with achieving good seals between metal alloy cladding, internal metal alloy coolant tubes, and the cermet fuel material.

• Insufficient permeation of alloy-clad material into the cermet during autoclaving, leaving weaknesses that developed into voids.

• Fission product damage to the cermet, and eventually the cladding material, caused by accumulated buildup of pressure, lattice stresses, and dislocation weaknesses under irradiation.

Sintering to lower theoretical densities of 84–90% created a significant improvement in sample performance. An increase in burnup capability (i.e., fissions/cm$^3$) by almost a factor of 10 was achieved by simply giving the fission products additional room for expansion without exerting stresses in excess of the tungsten matrix capability at elevated temperatures.

2.3 Argonne national laboratory nuclear rocket engine research and development program

The ANL nuclear rocket program focused on developing two reference reactor designs; the ANL200 and ANL2000 reactors were 200 MWe and 2000 MWe fast spectrum thermal propulsion systems that were designed to produce 44.5 kN (10,000 lbf) and 445 kN (100,000 lbf) of thrust.

Most of the ANL program’s work was focused on design and testing of the ANL2000 system. The reactor consisted of an array of 163 hexagonal fuel elements that were assembled into an approximately cylindrical core with a diameter of 66 cm (26 in). The fuel elements were made from a 93% enriched tungsten-urania
cermet fuel that was clad with 0.76 mm (0.03 in) of a tungsten-rhenium (W-25Re) alloy. The elements had a total length of 130 cm (51.56 in) and a fueled length of 87 cm (34.25 in). The core was supported from an Inconel Inco-718 grid plate that was bolted to the reactor vessel at the cold end of the core, and a cylindrical beryllium-oxide axial reflector containing 12 control drums was mounted at the inlet end of the core. A preheater consisting of stainless steel-UO$_2$ fuel elements at the inlet side of the reactor was also included in the reactor design.

The ANL2000 development program's performance goals were to reach a fuel temperature of at least 2770 K in order to produce an Isp of 821–832 s, achieve 10 hours of operation with at least 25 thermal cycles, and limit fuel loss to less than 1%. All of the program's goals were achieved before the program was terminated; however, neither of the ANL program's reference reactor designs were built or tested before the program was cancelled.

The primary fuel evaluated under the ANL program was UO$_2$ embedded in a tungsten matrix. The fuel choice was similar to the GE-710 program, but gadolinia was used to stabilize the ANL fuel, in contrast to the ThO$_2$ that was used in the GE-710 program. Three fuel fabrication methods were investigated under the program: cold pressing and sintering of W-UO$_2$ wafers, isostatic sintering of long fuel elements, and hot pneumatic compaction.

The cold pressing and sintering technique led to fabrication of approximately 6.3 mm (0.02 in) thick W-UO$_2$ wafers that were stacked to form a fuel column. Fuel grading was used in the stacks to optimize physics and thermodynamics of the core. The fuel fabrication method required a high strength cladding since the cladding provided structural support. The isostatic sintering method allowed for single-step fabrication of fuel elements that were approximately 45.7 cm (18 in) long. The process minimized concerns over coolant channel alignment tolerances because individual fuel wafers did not have to be stacked to form an element. Finally, the hot pneumatic pressing method was used to demonstrate the fabrication of fuel formed from UO$_2$ fuel kernels that were CVD coated with tungsten. A fuel loading of 60 vol% of 93% enriched UO$_2$ inside a W or W-Re matrix was used for all of the program's fuel samples.

Similar to the GE-710 program, stabilizers were added to the ANL program fuels to inhibit UO$_2$ dissociation, but the stabilizers investigated under the ANL program included gadolinium (GdO$_{1.5}$), dysprosium (DyO$_{1.5}$), yttrium (Y$_{1.5}$), and MoO$_3$. Ten mole percent of stabilizer was added to the UO$_2$ for all investigations.

The fuel fabrication process that gave the best results was a powder metallurgical process that produced near net shape fuels with cold isostatic pressures, followed by sintering at approximately 1500 K and chemical vapor deposition (CVD) of cladding on the coolant channels, even though deposition of uniform CVD coatings was difficult in the 1960s. The gadolinia stabilized fuel showed excellent retenivity at 2770 K for up to 45 hours and 180 cycles in non-nuclear tests performed in two hydrogen loops. Other tests showed that flowing hydrogen at temperatures exceeding 2700 K had essentially no impact on fuel loss rates.

Induction brazing was investigated by the ANL program as a means for joining fuel sections. A Zr-Mo braze with a melt temperature above 1973 K was the most successful; however gas generated during brazing made it almost impossible to fabricate a leak-free joint. The problem was overcome by immersing fuel sections in liquid nitrogen with the section to be brazed left above the liquid pool. Brazing was carried out in five sections to avoid allowing the fuel section to reach a temperature where volatilization of impurities could occur.

High-temperature refractory brazing techniques were also developed under the ANL program. Solid-state diffusion bonding of W-25Re alloys using nickel as an interleaf material that forms an Ni-W-Re ternary has been demonstrated at
temperatures as low as 1173 K (although a temperature of 1773 K is required to produce sufficiently strong bonding). Brazing of refractory metals is generally undesirable due to recrystallization of microstructures produced in the joint, but solid-state diffusion bonding avoids recrystallization through the use of low temperatures. Nickel may be an undesirable interleaf material for high-temperature NTP materials, but other interleaf materials may be identified with further investigation [11].

Nuclear tests on the ANL cermet samples were performed in the Transient Reactor Test (TREAT) facility at INL. Eight cermet specimens, each with seven coolant channels and vapor-deposited tungsten cladding, were tested in the TREAT experiments. The test durations were typically 200–430 ms, although two samples were subjected to flat-top transients lasting 2–3 s. One of the tests failed, as fuel material was ejected from the sample, and the failure was attributed to fabrication issues, particularly tungsten coating thickness irregularities. The last two samples evaluated in the campaign were subjected to multiple transients at heating rates up to 16,000°C/s, a maximum temperature of 2870 K, and a power density of 30 MW/l. These samples showed no evidence of damage [7].

2.4 Space Nuclear Thermal Propulsion research and development program

The goal of fuel development under the Space Nuclear Thermal Propulsion (SNTP) Program was to develop a coated nuclear fuel particle with a diameter of approximately 500 μm that would support a mixed mean hydrogen exhaust temperature of 3000 K when incorporated into a particle bed reactor (PBR) [12]. The requirement gave a maximum fuel temperature target of 3100–3500 K based on a power density of 40 MW/l, whereas the maximum fuel temperature demonstrated during the Rover/NERVA program was in the range of 2400–2600 K.

The particle bed reactor concept developed by Dr. James Powell and his team at Brookhaven National Laboratory (BNL) caught the attention of SDI program managers as a possible power source for a rapid intercept vehicle that could destroy ballistic missiles, because it had the potential to overcome limitations associated with high-power production. Interest in the PBR technology led to the creation of the Timberwind program in 1987 and creation of the SNTP program in 1991, after Timberwind was declassified and transferred to the US Air Force.

The PBR fuel element designed for the SNTP program consisted of a large number of UC$_2$ fuel particles packed between two porous cylinders called frits. The fuel elements were housed inside cylindrical moderator blocks made of beryllium or lithium hydride that slowed the reactor’s neutrons down to thermal energies that could sustain a fission chain reaction. Hydrogen served as both a coolant and propellant for the SNTP engine as it moved through the cold frit located on the outside of the fuel elements, flowed through the element particle beds to remove heat produced by the fission reaction, and then exited the fuel through the inner hot frit. The hydrogen then flowed axially down an annular channel located at the center of each of the core’s fuel elements and exited the core before expanding through the engine nozzle to produce thrust.

The PBR concept promised significant reductions in system mass over solid core reactors due to the 20-fold increase in heat transfer surface area of the particle fuel elements compared to the prismatic fuel used in the Rover/NERVA program. PBRs also had a lower core pressure drop due to the shorter flow paths through the pebble beds. The small size of the particles helped to prevent cracking, because thermal gradients across the particles are relatively low, but the coatings used on the particles were found to be prone to high-temperature vaporization that was made worse by the high surface area to volume ratio of the particles.
The SNTP program began working on the development of coated fuel particles based on the HTGR Program fuel design. These particles were known as the program’s baseline fuel. The baseline fuel development included the production of uranium-bearing fuel kernels using the internal gelation process. The fuel kernels were covered by pyrolytic carbon using chemical vapor deposition in a fluidized bed.

Babcock and Wilcox Inc. (B&W) developed the ability to produce ZrC outer coatings on microparticles with the assistance of LANL and General Atomics. B&W produced fuel particles consisting of UC\textsubscript{2-x} kernels coated with two layers of pyrocarbon and an outer layer of ZrC that supported the Particle bed reactor Integral Performance Element (PIPE) experiments that were performed in 1988 and 1989. The first pyrocarbon layer in the fuel particles was a porous layer that accommodated the mismatch in thermal coefficient of expansion between the fuel kernel and the outer ZrC layer. The second layer was dense pyrocarbon that protected the fuel kernel from attack by the halides used in the CVD process. The outer ZrC layer was used to delay corrosion of the fuel kernel after it was exposed to hydrogen propellant.

More than 200,000 particles were tested in Sandia National Laboratory (SNL) Annular Core Research Reactor (ACRR) in four particle nuclear tests (PNT) [12]. Fuel temperatures achieved during the tests ranged from 1800 to 3000 K, and testing times ranged from 100 to 600 s. Baseline UC\textsubscript{2-x} fuel kernel performance is limited by its melting temperature of 2700–2800 K, but the PNT tests showed that the melting temperature of fabricated UC\textsubscript{2-x} kernels was actually closer to 2500 K. Molten UC\textsubscript{2-x} dissolved the particle carbon layers and attacked the ZrC outer layer during the tests, and a complete particle failure occurred about 5 min after kernel melting. It is possible that increasing the graphite layer thickness would delay the time to failure, but the increased particle size might weaken any fuel matrix that was used to contain the particles, so testing of increased graphite layer thicknesses was not performed by the SNTP program.

The program pursued a dual fuel development path once it became clear that coated UC\textsubscript{2-x} kernels would not meet the program’s temperature requirements. Under the dual-path effort, BNL investigated the development of an infiltrated kernel (IK) fuel, and B&W investigated mixed-carbide fuel particles. BNL postulated that IK fuel could be formed when molten UC\textsubscript{2-x} distributes uniformly through a porous graphite matrix. The laboratory’s scientists reasoned that the molten uranium ceramic could be held within the graphite’s pores and protected from hydrogen corrosion by an appropriate high-temperature outer layer, since UC\textsubscript{2-x} is thermodynamically stable with respect to graphite and does not react with it even after melting. BNL demonstrated in 1992 at laboratory scale that molten UC\textsubscript{2} could be infiltrated into porous graphite coupons to the desired uranium density and that spherical IK particles could be fabricated. The demonstration also showed that pyrolytic layers used in the baseline fuel design are unnecessary in the BNL IK fuel, so IK fuel has a higher uranium density and smaller particle size than the baseline SNTP fuel.

The B&W mixed-carbide fuel design developed under SNTP was based on investigations that were performed at the end of the Rover/NERVA program. The fuel was formed as a mixture of refractory carbides such as ZrC, NbC, TaC, HfC, and UC. Uranium carbide has a theoretical melting temperature of 2798 K, but the refractory metal carbides have melting temperatures ranging from approximately 3700 K for ZrC to greater than 4200 K for TaC and HfC. Tantalum and Hf have relatively high neutron absorption cross sections, so only ternary mixtures of U-Zr-C and U-Nb-C were considered by the B&W fuel development program.

The diagram shown in Figure 2 is an example of phase relationships for a mixed carbon fuel [13]. As illustrated in the figure, the melting temperature of mixed-carbide fuels decreases with increasing uranium content. The necessary uranium content for SNTP fuel was determined by fuel criticality conditions, and the B&W research
identified a minimum required uranium mole fraction of 0.15, which equated to a melting temperature of approximately 3200 K. By the end of 1992, B&W measured the melting temperature of U-Zr-C as a function of uranium content; measured the plasticity of ZrC, NbC, and U-Zr-C at 3200 K; and produced a small amount of NbC-coated U-Zr-C kernels using an internal gelation manufacturing process and CVD coating.

Overall accomplishments of the SNTP program included:

- Acquisition of technology and equipment that allowed production of nuclear fuel microparticles using the internal gelation process and coating of the particles with pyrolytic carbon and refractory metal layers using CVD
- Production of baseline fuel particles that supported radiation and non-radiation testing
- Development of a laboratory process for infiltrating porous graphite with uranium to produce infiltrated kernel particles
- Modification of the internal gelation process that allowed for production of U-Zr-C particles

2.5 Russian space nuclear engine research and development

The Union of Soviet Socialist Republics (USSR) performed a significant amount of research and development on nuclear thermal propulsion fuels from the 1960s to the late 1980s. Reported work included:
• Fabrication of a large number of samples
• Non-nuclear hot hydrogen flow tests
• Individual fuel element tests under rapid transient conditions in the Impulse Graphite Reactor (IGR)
• Prototypic NTR operating condition testing in the IVG-1 and RA reactor

The USSR followed the NERVA program quite closely and chose to follow the mixed-carbide fuel path early in its fuel development program. (U, Zr) C fuel was used for the low-temperature portion of the USSR reactor design (i.e., propellant exit gas temperature \( \leq 2500 \) K), and (U, Zr, Nb) C was used for the high-temperature portion of the reactor core (i.e., propellant exit gas temperatures up to 3100 K). Some work using Ta and Hf in place of Nb was also reported. There were claims that Ta and Hf could produce 200 K higher fuel temperatures, but there was concern over the higher neutron capture cross sections of these elements compared to Nb. Finally, carbon nitride fuels were developed under the USSR program, primarily for use with ammonia propellants.

The USSR research program fabricated carbide fuels in a wide range of shapes, but the twisted ribbon geometry was the preferred fuel design. This geometry included long rods of fuel with many different cross-sectional shapes. The rods were twisted along their long axis and bundled together using wire wraps, or insertion into long canisters, to form fuel elements [14]. During the operation of the reactor, the propellant was directed through the bundles to transfer heat from the fuel. The twisted ribbon geometry provided a large surface area for heat transfer, and it could be fabricated in large volumes, although researchers from outside the USSR program were not allowed to observe the fabrication process.

Tests were performed on the USSR nuclear thermal rocket fuel design over a period of 19 years on approximately 1550 fuel assemblies. The testing program included seven full-core tests and approximately 160 transient tests that were performed at the IGR between 1962 and 1978 [7]. The highest reported hydrogen exit gas temperatures from testing performed during that period ranged from 2800 to 3300 K. Reported power densities were as high as 20 MW/l, and uranium loss estimates were as low as 0.5–1.0% based on reactivity loss measurements [15]. Very little postirradiation examination data on the fuel samples has been reported.

3. Carbon-based fuels and materials

The GE-710 and ANL programs were established as backups to the Rover/NERVA program. The choice of evaluating refractory metal-based fuels as a secondary fuel type to the Rover/NERVA graphite fuel research resulted from the greater experience base associated with graphite fuel, graphite’s low thermal neutron absorption cross section, and the greater fabricability associated with graphite fuels.

Graphite was first used in nuclear reactors as a moderator, and large bars of polycrystalline graphite were used in many early reactors. A halogen purification process was developed to produce the high-purity graphite needed for natural uranium-plutonium production piles. More recently, graphite has been used as a fuel particle coating and as a matrix for fuel particles in high-temperature reactors [16].

The term “graphite” refers to a wide range of materials made from carbon that have a variety of properties. For example, graphite can be used as both a thermal
conductor and a thermal insulator, it can be made in very dense and very light forms, and it can be highly anisotropic or isotropic. Graphite also has a wide variety of uses in nuclear reactor applications. It can serve as a high-purity neutron moderator, and it can be used in control rods and shielding with the addition of boron.

The variety of properties associated with nuclear grade graphite means that it can be difficult to obtain graphite that has specific properties within narrow limits that are consistent from batch to batch. New sources of graphite often have unknown property variations.

Graphite production processes are often proprietary, but the general method of manufacturing crystalline graphite includes [17]:

- Raw petroleum coke is calcined at 1300°C, milled, sized, and mixed at about 165°C with a coal-tar pitch.
- The mixture is cooled to 110°C and extruded.
- The extrusion is cooled to room temperature to form a “green body” and placed in a baking furnace supported by a permeable pack of sand and carbon.
- A large volume of gas evolves from pyrolysis of the pitch during baking to 800°C and the carbon body shrinks about 5 vol%.
- The material is then graphitized in an electric furnace at 2500–3000°C. Some further gas is vaporized during graphitization, but the principle physical change involves transformation from amorphous carbon into crystalline graphite.

There are many variations that can affect final material properties. For example, the baked carbon can be impregnated with pitch to increase density and strength, carbon black can be added to improve density and strength, and the graphitized body can be heated in a halogen-containing environment to remove trace impurities.

Pyrolitic carbon is made from decomposition of hydrocarbon gasses. For free-standing bodies, the carbon is usually deposited on a graphite substrate at temperatures from 1400°C to 2400°C. Material orientation, density, and other properties can be varied by changes in gas pressure, temperature, and other conditions. Subsequent heat treatment at higher temperatures can improve crystallinity, and small samples heat treated to 3000–3600°C (3273–3873 K) have shown electrical properties that are close to the properties of single crystals. Larger samples with near-single-crystal properties can be made by heating pyrolytic graphite above 2500°C (2773 K) under a compressive stress. Fuel particles are typically coated with pyrolytic carbon in a fluidized bed with the carbon coatings being applied to thicknesses of up to about 100 μm.

Carbon is a relatively light atom, so graphite is an efficient moderator. Slowing down power is the logarithmic energy change of a neutron when it collides with a moderator, and a nuclear graphite with a density of 1.65 g/cm³ has a slowing down power of 0.063 cm⁻¹/Å. Light water has the highest slowing down power of 1.5 cm⁻¹/Å, and only several other materials such as beryllium (Be), beryllium oxide (BeO), and deuterium oxide used in heavy water reactors (D₂O) have higher slowing down power than graphite. Graphite also absorbs fewer thermal neutrons than any other material except D₂O.

Graphite is relatively weak at low temperatures, with a compressive strength of only a few thousand psi. However, its high-temperature strength is very good compared to other materials. Graphite's strength increases with temperature and
reaches a maximum at about 2500°C (2773 K). A typical polycrystalline nuclear graphite with a tensile strength of 2000 psi at room temperature has a strength of about 4000 psi at 2500°C (2773 K). Graphite’s high-temperature strength, good nuclear properties, and low cost are the primary reasons for its extensive use in high-temperature gas-cooled and nuclear propulsion reactors.

Carbide fuels such as UC and UC$_2$ have advantages over more widely studied oxide fuels. The most important advantage is their higher thermal conductivity, which approaches the value found in metallic uranium. Higher thermal conductivity lowers peak centerline fuel temperatures, which in turn allows for higher linear heat generation and larger diameter fuel rods. Carbide fuels also have higher uranium densities than UO$_2$, which allows for design of more compact reactors [15].

Mixed carbides such as uranium-zirconium carbide solid solution ([U, Zr] C) fuels have higher melting temperatures than UC. Research into mixed-carbide fuel fabrication has taken place in the USA and former Soviet Union to support space nuclear power applications. Three major carbide fuel designs were investigated under Rover/NERVA:

- UC$_2$ particles with pyrolytic carbide coatings and dispersion in graphite
- Composites of (U, Zr) C and graphite with the carbide forming a continuous web structure within the fuel
- Solid solution (U, Zr) C

All of the fuel designs, except the solid solution design, used protective ZrC coatings. Only 28 solid solution fuel elements were tested under Rover/NERVA, so effectiveness of the fuel design was not fully evaluated.

A solid solution is formed when two metals are completely soluble in their liquid and solid states. Complete solubility means homogeneous mixtures of two or more kinds of atoms are formed in the solid state. The more abundant atomic form is referred to as the solvent, and the less abundant atomic form is referred to as the solute. For example, brass is a solid solution of copper (64%) and zinc (36%) so that copper is the solvent and zinc is the solute.

There are two types of solid solutions: substitutional solid solutions and interstitial solid solutions. Substitutional solid solutions, which can be either ordered or disordered, are formed when solvent atoms in the parent metal’s crystal structure are replaced by solute atoms. For example, copper atoms may substitute for zinc atoms without disturbing zinc metal’s face-centered cubic (FCC) crystal structure. For complete solid solubility, the two elements should have the same type of crystal structure, and for extensive solid solubility, the difference in atomic radii between the two elements should be less than 15% [18]. Solid solubility is favored when the two elements have lesser chemical affinity, since compounds form when chemical affinity is high. Generally, compounds that are separated in the periodic table have higher chemical affinity, so elements that are close together tend to form solid solutions.

In interstitial solid solutions, solute atoms enter holes in the solvent atom crystal structure in interstitial solid solutions. Atoms that have atomic radii less than 1 Å tend to form interstitial solid solutions. Carbon, nitrogen, oxygen, and hydrogen are example interstitial solid solution solutes. Intermetallic compounds are formed when one metal (e.g., magnesium) has chemical properties that are strongly metallic and another metal (e.g., antimony, tin, or bismuth) has chemical properties that are only weakly metallic. Intermetallic compounds have higher melting temperatures than either of their parent metals. The higher melting point indicates a strong chemical bond in the intermetallic compound.
Table 2 lists melting points and carbon to metal ratios (C/M) for several monocarbides that have been investigated for use in space reactors [19]. Solid solution carbides are expected to be able to operate for short periods of time at propellant exit temperatures as high as 3200 K and for many hours at exit temperatures of 2600–3000 K. The life-limiting phenomenon for the solid solutions appears to be vaporization at surface temperatures greater than 2900 K.

The highest melting temperatures for most monocarbides occur at C/M ratios that are less than one, and pseudo-binary and pseudo-ternary carbides have their highest melting temperatures for single-phase solid solutions. The melting point for single-phase solid solution carbides has been shown to be 100–700 K higher than the melting temperature for carbides that have formed a separate carbon phase (e.g., [U, Zr] C_6 + C) [20, 21]. Figure 3 shows the solidus curves for ternary mixed carbides of (U, Zr, Nb) C from [20]. This study showed higher melting temperatures for ternary mixtures than for binary carbides of ZrC or NbC with an equal amount of UC.

Carbon to metal ratios were carefully controlled in the Rover/NERVA Nuclear Furnace (NF-1) tests to prevent formation of second phases that significantly reduced melting temperatures of the carbide fuels. A C/M ratio of 0.88–0.95 was targeted for NF-1, (U, Zr) C fuel elements for a proposed maximum operating temperature of 3200 K [22].

<table>
<thead>
<tr>
<th>Binary alloy</th>
<th>Melting temperature (K)</th>
<th>Carbon to metal ratios</th>
</tr>
</thead>
<tbody>
<tr>
<td>NbC</td>
<td>3873 ± 25 K</td>
<td>0.79</td>
</tr>
<tr>
<td>TaC</td>
<td>4258 K</td>
<td>0.89</td>
</tr>
<tr>
<td>UC</td>
<td>2803 K</td>
<td>1.00</td>
</tr>
<tr>
<td>ZrC</td>
<td>3813 K</td>
<td>0.87</td>
</tr>
</tbody>
</table>

Table 2. Melting temperatures and carbon to metal ratios of various monocarbides [19].

Figure 3. Solidus curves for ternary mixed carbides of (U, Zr, Nb) C [20].
Fabrication of carbide fuel elements was completed in several steps under Rover/NERVA. First, a mixture of ZrC, UO$_2$, ZrO$_2$, graphite flour, and binder was prepared and extruded. Free carbon was removed from the extrusion by leaching with hot flowing hydrogen. The fuel elements were then impregnated with zirconium to varying degrees using a CVD process to produce a single-phase solid solution carbide element that was substoichiometric in carbon. Extrusion of these elements produced severe die wear because of the carbide content, so 19 mm (0.75 in) wide hexagonal elements containing 19 coolant channels could not be directly fabricated through extrusion. Instead, the hexagonal elements were manufactured by first extruding cylindrical fuel forms and machining them to a hexagonal geometry.

All three of the carbide fuel constituents can be mixed, cold pressed, and sintered to fabricate a fuel pin, but mixing all of the components at once makes it difficult to control C/M ratio and prevent the formation of a second carbon phase. Carbide particles also tend to be coarse and require long sintering times at high temperatures in order to produce a homogeneous material.

The major problem with the use of graphite and other carbon-based fuels (e.g., UC, UC$_2$, [U, Zr] C) in high-temperature space reactor applications is mass loss produced by a number of interrelated and competing physical processes [23]. These processes include the formation of carbon liquids, loss by vaporization, extensive creep, and corrosion during hydrogen exposure. Maximum mass losses typically occurred in moderate-temperature regions of the core (<2000 K). The amount of hydrogen corrosion that occurs is dependent on:

- Reactor operational duration
- Number of fuel duty cycles
- Local material temperatures
- Local hydrogen gas flow conditions
- Fuel location in the reactor
- Reactor power density
- Compatibility of the fuel and coatings

There are four major coupled reactions and/or healing processes associated with hydrogen corrosion:

- Exposure to hydrogen gas
- Nonuniform loading and/or cycling of the fuel
- Radiation exposure
- Creep

The first process is directly associated with chemical corrosion, while the remaining processes affect the amount of cracking that occurs in the fuel, which affects the fuel surface area that is exposed to hydrogen.

Carbon-based fuel materials can experience mass loss by two mechanisms when exposed to hot hydrogen: vaporization (or sublimation) of material constituents at
temperatures above carbide vaporization temperatures, and chemical reaction of carbon constituents with hydrogen to form hydrocarbon gas species such as methane (CH$_4$) and acetylene (C$_2$H$_2$). Vaporization occurs at varying rates in the moderate- to high-temperature regions of U-Zr-C fuel, but it is the predominant mass loss mechanism at temperatures greater than 2900 K. Little chemical reaction between hydrogen and carbide materials takes place at temperatures below approximately 1500 K, but chemical reaction losses predominate below the vaporization temperature of carbide materials (i.e., temperatures between 1500 K and 2900 K). The formation of CH$_4$ becomes increasingly unstable at low to moderate hydrogen pressures and temperatures greater than 1500 K, since C$_2$H$_2$ is the more stable compound under these conditions, but the opposite relationship is true for higher pressures [24].

Figure 4 shows the recession rate of U$_{0.1}$Zr$_{0.9}$C compared to other compounds as a function of temperature and illustrates the fact that the diffusion rates of carbon and uranium can be substantial at high temperatures. Changes in surface chemical conditions in U-Zr-C materials likely encourage the release of free carbon, since surface composition changes tend to enhance the diffusion of carbon and uranium to the fuel surface because of shifts in the U/Zr/C ratio. These changes were noted during start-up of the Rover/NERVA reactors and were determined to be a predominant contributor to corrosion mass loss. Corrosion mass loss may also degrade fuel surface properties so that particles, such as fuel grains and grain agglomerations, become loosened and erode into the hydrogen gas stream. The presence of hydrocarbons in the propellant stream tends to decrease the release rate of carbon from downstream fuel surfaces [26]. This effect may be a partial explanation for the lower corrosion rates that were observed in higher-temperature regions of the Rover/NERVA fuel elements. Another partial explanation for the lower corrosion rates may be the healing of surface defects due to material creep at high temperatures. This healing process may reduce hydrogen intrusion into the high temperature fuel regions, but the healing effect may be reduced by radiation damage. Radiation damage can also reduce the thermal conductivity of the fuel, which can produce locally high thermal stresses and
corresponding mismatches between stresses in fuel coatings and the fuel substrates. These mismatches in turn encourage the formation of surface coating cracks that enhance hydrogen penetration into the fuel and offset any beneficial effect produced by creep healing.

Another major hydrogen corrosion initiator is nonuniform loading and thermal cycling of the fuel. Nonuniform loadings and material expansion effects were considered to be a major cause of reduced corrosion in higher-temperature fuel regions due to closing of surface cracks [24]. Nonuniform mechanical loading can be produced by:

- The presence of preload stresses during fuel fabrication and application of material coatings. The Rover/NERVA reactors were often designed with tie tubes that had substantial preloads that offset the axial loads produced by high hydrogen pressure differences.
- The presence of nonuniform or unsteady pressures and nonuniform axial temperature distributions. Varying pressure profiles are always present during NTP transient operations, such as during reactor start-up and shutdown.
- Residual stresses produced by fuel cycling. For example, fuel can undergo creep at high temperatures, which may lead to high induced tensile and compressive stresses during fuel cooldown.

It was initially believed during the Rover/NERVA program that radiation effects would be minimal in carbon-based fuels because of carbon's resistance to radiation and the low operation time for NTP systems. This belief turned out to be unfounded because of the high-power densities that are required in NTP reactors. Post-test examinations of the Rover/NERVA reactors showed that radiation damage caused reductions in thermal conductivity and ductility, and these reductions caused cracking that allowed hydrogen to enter the fuel [22].

4. Future work

NASA is once again exploring the feasibility of building and operating nuclear fission systems for use in deep space science and exploration missions. The primary objective for the feasibility studies is to identify systems that can be used to support human missions to Mars, but missions to the outer solar system and beyond are also under consideration. The major barrier to demonstrating a high-performance nuclear propulsion system is developing a fuel that can survive the extreme operating conditions that will be required during space flight missions. The fuel operational characteristics that will need to be satisfied during reactor operations include:

- Minimizing high-temperature hydrogen corrosion
- Minimizing brittle fracture behavior at low temperatures
- Minimizing fuel creep and vaporization at high temperatures
- Minimizing radiation damage that impairs fuel performance
- Managing high transient thermal and mechanical stresses on the fuel during reactor start-up
• Rapid heat transfer from the fuel to the propellant

• Matching coefficients of thermal expansion for different materials used in the fuel to avoid fuel constituent separation during reactor operation

• High uranium loading to allow for use of low-enriched uranium fuel

• Low fuel and reactor system mass to minimize launch costs

• Limiting fuel dissociation and constituent migration during reactor operation
• Limiting cracking of fuel and coatings to minimize hydrogen ingress into the fuel during reactor operations.

As a result of these restrictive requirements, it is likely that whatever fuel is selected will have to operate close to its thermal and mechanical failure limits. There will be little margin for error in system operation, so a significant amount of research and testing will be needed before a safe and reliable system can be built and operated. The majority of future work associated with developing space reactor propulsion and power generation reactors will be associated with designing, building, and operating the equipment and experiments that will build on past testing programs and lead to fuel and reactor qualification and public acceptance.

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