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Neutronic effects of rhenium, gadolinia and uranium dioxide addition to a tungsten based fast spectrum space reactor

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ABSTRACT

The neutronic effects of rhenium (Re), gadolinia (Gd_2O_3) and uranium dioxide (UO_2) addition to a W–60 vol.% UO_2 fast spectrum fuel form were investigated relative to a prismatic reactor type using the MCNP5 Monte Carlo code with the ENDF/B-VII.1 cross-section database. The effects of rhenium addition to the cladding tubes and sleeves were studied over the range of 2–25 at.% as well as the corresponding effects on the reactor mass and control drum reactivity swing and neutron energy spectrum. The effects of rhenium addition to the tungsten matrix over the range of 1–5 at.% was also investigated. Lastly, the UO_2 concentration was varied from 55 to 60 vol.% UO_2 and the UO_2 was mixed with Gd_2O_3 at concentrations ranging from 2 to 10 mol.%. Each configuration was investigated for its effects on the control drum shut down margin both in a dry state and in a submerged state where the reactor was surrounded and flooded with fresh water. The high thermal spectrum absorption cross-section of tungsten, rhenium and gadolinium proved excellent in maintaining the reactor in a subcritical state when submerged in fresh water. However, increases in the concentration of rhenium and gadolinia to the matrix, cladding and fuel kernels were also accompanied by an increase in reactor mass. The control swing within the drums was not influenced by the material compositions; however, the increase in reactor size with varying UO_2 and Gd_2O_3 concentrations did prevent a large fraction of the neutron population from reaching the control drums. The decrease in neutron population with access to the reflector region containing the control drums caused a loss of reactivity swing in the drums by up to 2.8% $\Delta k/k$ ($4.36).

1. Introduction

Over the past 40 years there have been many attempts to develop and deploy space reactors fueled with tungsten based Ceramic–Metallic (CERMET) fuel elements (Rocket Program Terminal Report, 1966; High Temperature Gas Reactor Program Summary Report, 1973a; High Temperature Gas Reactor Program Summary Report, 1973b). The base constituents within CERMET fuels used for high temperature space operations are usually tungsten (W) and uranium dioxide (UO_2). There have been many development efforts to fabricate and test W–UO_2 fuel forms at the General Electric (GE) High Temperature Gas Reactor Program Summary Report, 1973a; High Temperature Gas Reactor Program Summary Report, 1973b, Argonne National Laboratory (ANL) Rocket Program Terminal Report, 1966 and National Aeronautics and Space Administration (NASA) Johnson et al., 1965; Marlow and Kaznoff, 1970; Lobsinger et al., 1966; Gedwill et al., 1965; Gripshover and Peterson, 1969; Takkunen, 1967; Sikora and Millunzi, 1968; Lamartine and Hoppe, 1965 as well as basic attempts at characterizing the neutronic effects of tungsten and rhenium additions to critical geometries (Webb and Charit, 2011). The traditional formula for fabricating W–UO_2 elements involves coating spherical UO_2 particles with tungsten to a predetermined tungsten volume fraction (MacInnis and Schulze, 1965; Angelo and Buden, 1985). Once the UO_2 particles are coated with tungsten, they are sintered into a highly dense state of the appropriate geometry by a process such as Hot Isostatic Pressing (HIP) Lamartine and Hoppe, 1965, Hot Uniaxial Pressing (HUP) Marlow and Kaznoff, 1970 or Pulsed Electric Current Sintering (PECS).

While tungsten and uranium dioxide are the primary constituents in W–UO_2 CERMET fuels, there are also other elements which can be added to the UO_2 fuel or the tungsten matrix to improve mechanical strength and high temperature fuel stability. Most nuclear propulsion reactors conceived for space flight require operational temperatures at or above 2700 K for up to 4 h of lifetime

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(Finseth, 1991; Takkunen et al., 1967). Uranium dioxide fuel has a very high vapor pressure and will vaporize if exposed to vacuum conditions at these temperatures. The coolant of choice for nuclear propulsion reactors is hydrogen, which is highly mobile and will migrate through tungsten by grain boundary diffusion (Lobsinger et al., 1966). Once the hydrogen reaches the UO_2 kernels it can reduce the UO_2 leading to the formation of free oxygen and liquid uranium, both of which have deleterious consequences for long term fuel stability. One option that has been successfully demonstrated to resist fuel vaporization and hydrogen reduction is the use of stabilizers such as CeO_2, Y_2O_3 and Gd_2O_3 (Bhattacharyya, 2001). These stabilizers develop a fluorite lattice structure with UO_2 raising the decomposition temperature with a slight decrease in the melting temperature. Of all stabilizers tested with W–UO_2 fuel, Gd_2O_3 (gadolinia) seemed to have the greatest effect in reducing fuel loss as a function of thermal cycles at temperatures above 2700 K (Klopp). The ANL rocket program conducted the most rigorous testing of Gd_2O_3 within the kernels at concentrations ranging from 2.5 to 10 mol.%. The optimum Gd_2O_3 concentration is very much dependent on the operational requirements of high temperatures and the need for many start-up-shut-down cycles. The GE program was able to manufacture and design reactors for UO_2 concentrations as high as 55 vol.%, whereas the ANL program was able to fabricate fuels at 60 vol.% UO_2.

The GE high temperature reactor program of the late 1960's and early 1970's investigated tungsten based CERMET fuels for use in nuclear propulsion and power production (High Temperature Gas Reactor Program Summary Report, 1973a; High Temperature Gas Reactor Program Summary Report, 1973b). The pre-conceptual design for a closed cycle power reactor proposed by GE was to operate at lower temperatures than were required for propulsion. The design temperatures ranged from 1900 K to 2400 K, which were also accompanied by higher levels of burnup over many months to years of continuous operations. The high values of burnup added another mechanical issue to the survivability of the fuel in the form of creep induced by gaseous fission products trapped in the lattice structure. One method of increasing the mechanical strength of the tungsten matrix was to alloy it with rhenium (Re). Rhenium has long been known to cause dramatic increases in the strength of tungsten while decreasing the ductile to brittle transition temperature (Klopp et al., 1965; Garfinkle et al., 1968; Conway and Flagella, 1968). While rhenium does increase the creep strength of tungsten at temperatures in the range of 1900 K to 2400 K, test data indicates that it actually decreases the creep strength of interest to propulsion reactors, which operate above 2400 K (Storms, 1987). Rhenium is also a neutron absorber which can cause radical increases in the critical dimensions of a reactor if used at high concentrations within the tungsten matrix. The General Electric company experimented with the use of solid solution W–Re alloys as a matrix material with Re concentrations as high as 5 at.%. Previous testing has indicated that the formation of liquid uranium in CERMET fuels containing rhenium can also lead to the formation of a URe_2 intermetallic compound, which melts at low temperatures and is accompanied by a large volume change (Bhattacharyya et al., 1057; Monte Carlo Team, 1987). Both the GE company and the ANL programs investigated the use of cladding materials that consisted of W–Re alloys with a Re concentration up to 25 at.%.

To date there is no comprehensive parameter survey which details the effects of varying UO_2, Gd_2O_3 and Re concentrations on a baseline space reactor design with respect to critical mass, reactor control, neutron energy spectrum and water submersion scenarios. This study uses the MCNP5 version 1.40 Monte Carlo computer code (King and El-Genk, 2005) with the ENDF/B-VII cross section database to investigate the neutronic effects of varying species loading on a pre-conceptual space reactor design.

2. Reactor physics analysis

The dimensions, mass, controllability and shutdown margin of any reactor is based on the fission, absorption and scattering cross-sections within the individual species present in the reactor. Traditional W–UO_2 CERMET reactor designs operate in the fast neutron energy spectrum where tungsten, rhenium, gadolinium and oxygen are relatively transparent to scattering events and are usually forward biased when a scattering event does occur. The lack of an appreciable scattering cross-section within the primary fuel nuclides leaves the absorption and fission cross-sections as the dominant drivers to neutronic performance.

Fig. 1 shows the energy dependent cross sections for U^{233}, U^{235} and Pu^{239} over the energy range of 0.0001 eV to 30 MeV, which were extracted from the ENDF/B-VII.1 cross section database provided by the National Nuclear Data Center.

There are many tradeoffs that can be made when determining what portion of the neutron energy spectrum one desires a space reactor to operate in. The choice of the average neutron energy can affect the reactor mass, moderator mass, mass of highly enriched uranium and the reactor physics parameters. However, operating a reactor in the fast neutron energy spectrum allows for the use of spectral shift absorbers which can help control the fission reactor in the event of an unintended excursion (Duderstadt and Hamilton, 1976).

If for some reason a reactor system finds itself surrounded by a high density, low molecular mass medium such as water, a series of unintended events may occur. First, the water molecules have a high lethargy for decrementing the neutron energy in each collision. On average a fast spectrum neutron need only undergo 16 collisions to become a thermal spectrum neutron, where its cross-section for fission is orders of magnitude higher (Lamarsh and Baratta, 2001; Hussey et al., 1965). Water molecules also have a high scattering cross section with an isotropic bias, which acts as a very good reflector to increase the neutron population within the fission core of a submerged reactor. The combination of the increased neutron population and radically increased fission cross-section can push an otherwise subcritical reactor into a critical excursion. The possibility of a water submersion scenario is particularly acute to the space reactor community since over 70% of the Earth's surface is covered by water. If a reactor system were to fall back to the surface of the Earth, it is likely that it may land in a submerged state.

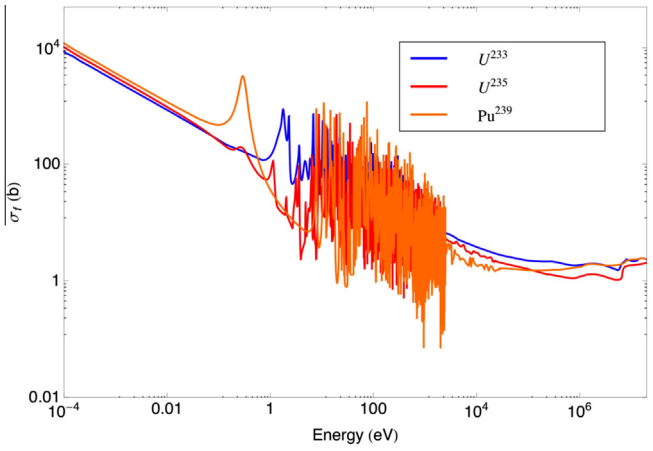

Fig. 1. Fission cross sections for uranium-233 (blue) uranium-235 (red) and plutonium-239 (orange), courtesy of the National Nuclear Data Center. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)