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Evaluation of uranium thorium and plutonium thorium fuel cycles in a very high temperature hybrid system

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ABSTRACT

In recent times, there is a renewed and additional interest in thorium because of its interesting benefits. Thorium fuel cycle is an attractive way to produce long term nuclear energy with low radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium or civilian plutonium. Th-based fuel cycles have intrinsic proliferation-resistance and thorium is 3–4 times more abundant than uranium. Therefore, thorium fuels can complement uranium fuels and ensure long term sustainability of nuclear power.

In this paper, the main advantages of the use of fuel cycles based on uranium-thorium and plutonium-thorium fuel mixtures are evaluated in a hybrid system to reach the deep burn of the fuel. To reach this goal, the preliminary conceptual design of a hybrid system composed of a critical reactor and two Accelerated Driven Systems, of the type of very high temperature pebble-bed systems, moderated by graphite and cooled by gas, is analyzed.

Uranium-thorium and plutonium-thorium once-through and two stages fuel cycles are evaluated. Several parameters describing fuel behaviour and minor actinide stockpile are compared for the analyzed cycles.

1. Introduction

Deep burn transmutation in Very High Temperature Reactors (VHTR) is based on using the neutron thermal spectrum to reach a high fuel burnup. Simplified comparisons made between capture and fission cross sections of fast and thermal neutrons concluded that transmutation using thermal reactors is not feasible. Nevertheless, more detailed studies carried out later proved that thermal neutrons are capable to transmute Minor Actinides (MA) in appropriate conditions (Venneri et al., 2001).

In the deep burn concept, transmutation of long lived wastes composed of transuranic elements from the nuclear reactors and almost the whole destruction of the materials useful for the nuclear weapons fabrication (plutonium isotopes), is obtained with only once-through reprocessing cycle (Baxter and Rodriguez, 2001).

One of the key benefits of the proposed system in this paper is the utilization of the fuel confined into TRISO (Tri-structural isotropic) particles. Graphite coated fuel particles present some attractive advantages for deep burn transmutation as they have high resistance to irradiation damages, mechanical stress, and have high melting points, enabling to reach a high fuel burnup (Maki et al., 2007). Such performance is central to the deep burn transmutation concept (Fokuda et al., 1995). The different TRISO particle's layers constitute excellent fission product and radionuclide barriers in the geological repository (US Nuclear Regulatory Commission, 2004). Ceramic fuel particles also keep impenetrable to humidity longer than current metallic containers destined to preserve the spent fuel of conventional nuclear reactors. These features make TRISO particles a robust and attractive waste container.

VHTRs belong to next nuclear plants generation, the Generation IV (U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, 2002). VHTRs are expected to have attractive features like low electricity generation costs and short construction periods. The development of this technology is based on the experience from High Temperature Gas Reactors (HTGRs) like Dragon Peach Bottom, from England, and German Arbeitsgeminshaft Versuchsreaktor (AVR) and Thorium High Temperature Reactor (THTR), from Germany. The above mentioned are experimental reactors that were built in the 60's to demonstrate their viability for electricity and heat cogeneration. They enabled to reach high coolant temperatures at the core's outlet. Current

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projects, i.e. HTTR-2000, from Japan and HTR-10, from China, which became operatives in the years 2000 and 2003 respectively, enable to reach coolant outlet temperatures higher than 950°C (Talamo and Gudowski, 2005).

Taking advantage of the high coolant's outlet temperatures in a VHTR, it is possible to produce hydrogen from water and heat utilizing the Iodine–Sulfur thermochemical process, or from water, heat and natural gas applying steam reforming. Electricity production with a high thermodynamic efficiency is also possible in a direct cycle with a gas turbine. The heat extracted from the reactor is also useful in refineries, petrochemical and metallurgic industries (Abánades and García, 2009).

Because of its constructive characteristics, TRISO fuel can reach a high burnup as mentioned above. It could work in a VHTR and later in a Very High Temperature Accelerator Driven System (ADS). This strategy would enable to reach the allowed high burnup for this kind of fuel with no structure damages due to irradiation or high temperatures.

An ADS is composed of a heavy particles accelerator, where high energy protons are generated. Protons react with a spallation target composed of heavy materials to produce neutrons. The spallation target is located at the centre of a subcritical core which contains the nuclear fuel. The nuclear fuel may be composed of traditional nuclear fuel, transuranic elements, long lived fission products to transmute them into short lived isotopes, or uranium-thorium (U + Th) and plutonium-thorium (Pu + Th) mixtures, as in the present study. Fissile elements are disposed in a way that nuclear chain reaction cannot be sustained without a neutron source (in this case are the neutrons from spallation reactions). Neutrons and charged particles are generated from spallation reactions when the accelerated protons hit the target. These neutrons are tightly linked to the kinetic behaviour of the system, like delayed neutrons in critical reactors. Therefore, the low fraction of delayed neutrons of the spent fuel isotopes loses importance in the chain reaction control, which is determined by the neutron source and the accelerator.

Neutrons produced from spallation reactions in the target and multiplied by fission neutrons in the nuclear fuel, can transmute transuranic elements and long lived fission products into stable elements. As a result of fission reactions that take place in the transmutation processes, large quantities of heat are produced and can be transformed into electricity.

The calculation methods used in this work are based on probabilistic computational modelling. The principal tool for particle transport and fuel burnup calculations was MCNPX code, version 2.6e (McKinney et al., 2007). This software was created in 1994 by Los Alamos National Laboratory. It is based on the Monte Carlo probabilistic method for computational modelling of time dependent transport of many kinds of particles, with a wide range of energies, in much different geometries. Version 2.6e incorporates new capabilities with respect to previous versions. Some of them were used in the present study, such as CINDER90 code for fuel burnup calculation (Wilson et al., 1999). The available library in XSDIR, ENDF/B VI.2, was used.

In the present work, we analize the feasibility of once-through cycles based on U + Th or Pu + Th fuel mixtures, with two stages: the first in a very high temperature pebble-bed graphite-gas critical reactor, and the second in two ADSs with similar characteristics of the reactor. To reach this goal, we propose a preliminary conceptual design of the hybrid system composed of the mentioned critical reactor and the two ADSs. We evaluated the advantages of the proposed fuel cycles and compared them with traditional cycles in terms of those parameters that describe the behaviour of fissile fuel and MA stockpile.

The characteristics and advantages of thorium based fuel cycles are introduced in Section 2. In Section 3 a once-through and two-stages (a VHTR and two ADSs) fuel cycle in a VHTR hybrid system is shown and described. The conceptual design of a very high temperature hybrid system for a U + Th deep burn cycle is made in Section 4. Some of the analyzed items are: the optimization of the physical parameters, fuel burnup, variation of the isotopic composition and the radiotoxicity of the long lived wastes. By the end of the section a comparison between a U + Th cycle and an equivalent uranium cycle is presented. In Section 5 the possibilities of Pu + Th cycles in the very high temperature hybrid system are studied. Finally, some conclusions are discussed in Section 6.

2. Uranium + thorium cycle

In uranium-thorium fuel cycle, reactor's criticality is mainly sustained by three isotopes: U^{235}, which represents a certain percent of the fresh uranium; U^{233}, which is produced by transmutation of the fertile isotope Th^{232}, and Pu^{239}, produced by transmutation of the fertile isotope U^{238}. In addition, Pu^{241}, formed by neutronic capture of Pu^{240}, modestly contributes to the energy released by fission during the fuel cycle, but its contribution to the final inventory of fissile isotopes is poor because of its short half life, as it is converted almost completely into Am^{241} during the cooling time of the wasted fuel.

In order to compensate the U^{235} depletion by means of U^{233} and Pu^{239} breeding, the amount of fertile nuclides must be greater than the amount of U^{235} due to the small capture cross section of the fertile nuclides in the neutron thermal energy range compared to the capture cross section of the U^{235}. At the same time, the amount of U^{235} must be large enough to set the criticality of the reactor (Talamo and Gudowski, 2005).

In nature, thorium exists as a natural radioactive metallic element. The general certain about thorium stockpile and allocation is quite limited because the global demand of thorium has been rather insignificant.

A number of thorium-based fuel cycle benefits follow (International Atomic Energy Agency, 2005):

- It is 3–4 times as abundant as uranium, is widely distributed in the earth crust and is easily commercially exploitable.
- Thorium fuel cycle is a very attractive way to produce long term nuclear energy with low level radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium or civilian plutonium.
- The absorption cross section of Th^{232} for thermal neutrons (7.4 b) is nearly three times that of U^{238} (2.7 b). Hence, the conversion of Th^{232} into U^{233} is theoretically higher than the currently used transformation of U^{238} into Pu^{239}. Therefore, thorium could be a better fertile material than uranium in a thermal neutron spectrum.
- For the fissile nuclide U^{233}, the number of fast neutrons produced per thermal neutron absorbed is greater than that for U^{235} and Pu^{239}. Thus, in contrast to the U^{238}–Pu^{239} cycle, in which breeding can be obtained only with fast neutron spectrum, in the Th^{232}–U^{233} fuel cycle breeding can be obtained with fast, epithermal or thermal spectra.
- The U + Th fuel cycle produces a lower amount of Pu and MA (Np, Am and Cm) than the U + Pu fuel cycle, which contributes to minimize the radiotoxicity associated to the spent fuel. Consequently, a thorium based fuel cycle produces less hazardous waste than the U–Pu fuel cycle used in current LWRs.

Some challenges associated to the use of thorium based fuels are under investigation (International Atomic Energy Agency, 2005).