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The TRANSURANUS burn-up model for thorium fuels under LWR conditions

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ABSTRACT

The Transuranus Burn-up (TUBRNP) model for (Th,Pu) O_2 and (Th,U) O_2 fuels is described and validated. To this end, the depletion equations of the nuclides ^{232}Th, ^{233}U and ^{234}U were included as well as the breeding of ^{235}U through a neutron capture in ^{234}U. The Monte Carlo code Serpent was utilized to derive the one-group effective cross sections and the radial power-shape functions that account for the resonance absorption in the epithermal range of neutron energies in ^{232}Th and ^{240}Pu. Finally, TUBRNP is completed by extending the fission yields of ^{233}U for the most representative isotopes of Nd, Xe, Kr and Cs. The validation of TUBRNP for (Th,Pu) O_2 type fuels was carried out in two steps. A comparison of the normalised radial distributed concentrations of Th, U, Pu, Nd, Xe and Cs between TUBRNP, Serpent and Electron probe microanalysis (EPMA) data points measured on a sample from a rodlet irradiated at Kernkraftwerk Obrigheim (KWO) allows checking the correctness in the derivation of the radial functions. In a second step, a comparison of the radially averaged values of the same elements between TUBRNP, Serpent, EPMA and benchmark codes under same conditions indicates the agreement in the computation of the one-group cross sections. For (Th,U) O_2 fuels isotopic compositions measured in a rod segment from the Light Water Breeder Reactor (LWBR) Shippingport irradiation programme were employed to validate the calculation of the 1-group cross sections in TUBRNP for this type of fuel.

1. Introduction

The interest in thorium as nuclear fuel has risen again to supply a clean, safe and cheap power that can cope with the environmental challenges and the increasing electricity demand. As shown in Fig. 1, the utilization of thorium lies in the breeding of ^{233}U through neutron capture of ^{232}Th and two beta decays of nuclides ^{233}Th and ^{233}Pa. In comparison to the conventional fissile actinides ^{235}U and ^{239}Pu, ^{233}U presents a better fission factor (η) in the thermal range (Lung, 1997) that yields a better neutron economy in the reactor. Moreover, the necessity of combining thorium with a fissile element in order to start up the cycle will facilitate non-proliferation issues by incinerating, for example, the existing stockpile of Pu from Mixed Oxides (MOX) assemblies or from atomic weapons.

In contrast, the presence of ^{233}Pa in the thorium cycle can be seen as a disadvantage due to its half-life (27 days), that in combination with a considerable radiative capture cross section at 0.0253 eV (2200 m/s) (40 barns Lung, 1997; Belle and Berman, 1984) may delay the breeding of ^{233}U. Furthermore, the decay of ^{233}Pa to ^{233}U inserts a positive reactivity in the reactor that could affect its shutdown margin. However, the largest drawback found in the thorium cycle is the breeding of ^{232}U through a (n,2n) reaction from ^{233}U. ^{232}U is at the head of a decay chain that has high-energy gamma emitters, such as ^{208}Tl and ^{212}Bi, with 2.6 and up to 1.8 MeV gamma energy decay and very short half-lifes of 3.1 and 60.6 min respectively (Lung, 1997) which requires a special handling procedures of the irradiated fuels.

Thorium-based fuels have been tested and studied worldwide along the nuclear industry history (Lung, 1997; Belle and Berman, 1984; Vijayan et al., 2017; Kannan and Krishnani, 2013; Ganesan, 2016). The first use of thorium reported was in the USA at the Borax IV & Elk River Reactors from 1963 to 1968, where (Th,^{235}U) O_2 pellets were mounted in Boiling Water Reactor (BWR) assemblies (IAEA, 2005; Das and Bharadwaj, 2013). Later in 1977 at the Shippingport power station in Pennsylvania a full Light Water Breeder core of ThO_2-(Th,U) O_2 was operated until its decommissioning in 1982 (Olson et al., 2002).

More recently, the European Commission (EC) has funded the projects Thorium Cycle (TC) (Somers et al., 2013) and Oxide Fuels: Microstructure and Composition Variations (OMICO) (Verwerft et al., 2007; Boer et al., 2016) in which (Th,Pu) O_2 test rodlets were irradiated under Pressurized Water Reactor (PWR) conditions. In Norway the

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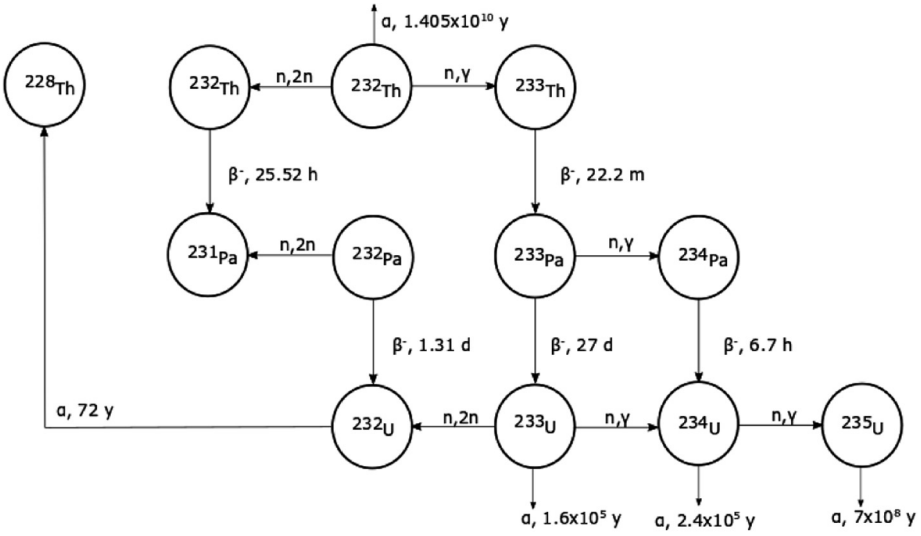

Fig. 1. Thorium cycle.

company ThorEnergy recently launched the program "Seven-Thirty" that aims at the manufacture and posterior test of (Th,U) O_2 and (Th,Pu) O_2 pins in the Organisation for Economic Co-operation and Development (OECD) Halden Reactor (Bjork et al., 2015).

The simulation of the nuclear fuel behaviour is important for licensing purposes and for interpreting experiments with new fuels. It involves the assessment of temperature distributions and stresses, as well as the evolution of the various fissile and fertile isotopes. Therefore, one of the first steps in describing fuel rod behaviour is to calculate the power distribution, i.e. at each fuel position the fraction of fissile material burnt or local burnup, the conversion of ^{232}Th to ^{233}U and the subsequent buildup and fission of the higher U isotopes. Indeed, the power density provides the source term for the temperature calculation, affecting most mechanisms in the code, as well as the source term for the radioactive fission products. The equations used constitute the so-called burnup models. The corresponding module included in the Transuranus fuel rod performance code is called TUBRNP and is currently used for oxide fuels with U and Pu. The standalone TUBRNP model is based on the earlier nuclide depletion model RADAR (Palmer et al., 1983). TUBRNP is employed in other fuel performance codes like Frapcon (Geelhood et al., 2011) and Falcon (Yagnik, 2002) or used as reference in the development of newer codes like BISON (Hales et al., 2013), AMP (Clarno et al., 2012), and Dionisio (Soba et al., 2013).

The current paper describes the extension of the TUBRNP model to thorium containing oxide fuels. In the next section, the set of depletion equations is extended to include the most important nuclides in the thorium cycle. These equations are solved incrementally by using the one-group effective cross sections for neutron capture and neutron-induced fission. The neutron flux is calculated from the analytical solution of the neutron diffusion equation. In addition, the continuous-energy Monte Carlo code Serpent (Leppanen, 2013) was run to compute the one-energy-group effective cross sections for representative fuel assemblies from the Shippingport programme and TC at KWO for (Th,U) O_2 and (Th,Pu) O_2 fuel cases respectively. In the third section of the paper, the selected experimental data for the validation is described. The validation of TUBRNP for both fuel types relies on the comparison of TUBRNP predictions with experimental data obtained from samples extracted from those previously mentioned thorium programmes. In the fourth section the results of this comparison are presented and discussed while in the last section the outcome and perspectives are summarised.

2. Extension of TUBRNP

In the standalone TUBRNP model (Schubert et al., 2008; Lassmann et al., 1994; Lassmann et al., 1998) the local power density generated within the fuel pellet, $q'''(r)$ (Eq. (1)) is proportional to the one-energy-group effective neutron-induced fission cross sections $\sigma_{f,k}$, the most relevant nuclides concentration $N_k(r)$ in the thorium fuel cycle and the thermal neutron flux, $\phi(r)$.

$$q'''(r) \propto \sum_k \sigma_{f,k} N_k(r) \phi(r) \tag{1}$$

In order to determine the radial shape of the flux, thermal diffusion theory can be applied that results in modified Bessel functions (Eq. (2)) (for further details see Schubert et al., 2008; Lassmann et al., 1994; Lassmann et al., 1998; Cavia et al., 2015)

$$\phi(r) \propto I_0(\kappa r) \tag{2}$$

The inverse diffusion length κ is derived from the macroscopic total neutron absorption and transport cross sections ($\Sigma_{a,tot}$ and Σ_{tr}) as follows:

$$\kappa = \sqrt{\frac{\Sigma_{a,tot}}{D}}; D = \frac{1}{3\Sigma_{tr}} \tag{3}$$

Here the total neutron absorption cross section is approximated by the microscopic absorption cross sections in the thermal range of neutrons:

$$\Sigma_{a,tot} = \sum_k \sigma_{a,th,k} N_k(r) \tag{4}$$

The set of depletion equations in TUBRNP for thorium fuels is extended to include the nuclides ^{232}Th, ^{233}U and ^{234}U and the breeding of ^{235}U through neutron capture in ^{234}U. In this first thorium version of TUBRNP the generation of ^{233}U is approximated to an immediate radiative capture in ^{232}Th by neglecting the formation of ^{233}Th and ^{233}Pa. The generation of ^{232}U through a (n,2n) reaction is also disregarded due to the small cross section of this reaction (Lung, 1997).

$$\begin{aligned}
\frac{dN_{Th232}(r)}{dbu} &= -A\sigma_{a,Th232} N_{Th232}(r) f_{Th232}(r) \\
\frac{dN_{U233}(r)}{dbu} &= -A(\sigma_{a,U233} N_{U233}(r) - \sigma_{c,Th232} N_{Th232}(r) f_{Th232}(r)) \\
\frac{dN_{U234}(r)}{dbu} &= -A(\sigma_{a,U234} N_{U234}(r) - \sigma_{c,U233} N_{U233}(r)) \\
\frac{dN_{U235}(r)}{dbu} &= -A(\sigma_{a,U235} N_{U235}(r) - \sigma_{c,U234} N_{U234}(r))
\end{aligned} \tag{5}$$

N_k represents the concentration of the nuclide k and A is a conversion factor that transforms a time step into a burn-up step (for further details see Lassmann et al., 1994). $\sigma_{a,k}$ and $\sigma_{c,k}$ are the spectrum-averaged absorption and radiative capture microscopic cross sections which are geometrically and burn-up averaged. $f_{Th232}(r)$ is a radial-