Annals of Nuclear Energy 99 (2017) 399–409

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

The experimental determination of the $^{238}U(n,\gamma)$ and total fission reaction rates along the pellet radius of the IPEN/MB-01 reactor

Luís Felipe Liambos Mura, Adimir dos Santos *, Douglas Borges Domingos, Pedro Carlos Russo Rossi, Rogério Jerez

Instituto de Pesquisas Energéticas e Nucleares, Av. Lineu Prestes 2242, CEP: 05508-000 São Paulo, SP, Brazil

ARTICLE INFO

Article history:
Received 29 June 2016
Received in revised form 23 September 2016
Accepted 27 September 2016
Available online 20 October 2016

Keywords:
Spectral indices
IPEN/MB-01 reactor
^{238}U and ^{235}U reaction rates
Intra-pellet measurements

ABSTRACT

This work presents a new experimental approach to determine experimentally the reaction rate along the pellet radius of a research reactor facility. The methodology employs concentric hollow cylindrical lead collimators together with gamma-ray spectrometry in a special kind of HPGe detector for the discrimination of very low gamma energy emitted by ^{239}Np and ^{99}Mo. The ^{238}U neutron capture and total fission rates were inferred, respectively, from the 106.2 keV gamma emitted by ^{239}Np and from the 140.51 keV gamma-ray emitted by ^{99}Mo. These lower gamma-ray energies allow the thickness of the lead collimator to be small enough so that the correction factors applied to the procedure were minimized. This kind somewhat challenging experiment was successfully performed at the IPEN/MB-01 research reactor facility. The experiments are claimed to be well-defined, and they are suitable for a benchmark. The measured values of the total fission rates are mainly due to the thermal fissions in ^{235}U since in the IPEN/MB-01 reactor nearly 85% of the total fissions occur in the thermal neutron energy region. The theoretical analyses were performed using MCNP-5 together with the ENDF/B-VII.0 library. The analyses reveal a very good agreement between the calculated and experimental results for the ^{238}U epithermal neutron capture reaction rates. However, the same can not be said for the thermal reaction rates which show discrepancies both in magnitude as well as in the shape of the attenuation of the reaction rates inside of the fuel pellet. The suspected reason for these discrepancies is the shape of the ^{235}U cross sections below 0.3 eV which might be different from that adopted in the ENDF/B-VII.0 library.

1. Introduction

In the fuel burnup evaluation process it is crucial to take into account the radial distribution of power and temperature inside the fuel pellet. This is usually achieved by dividing the fuel pellet into concentric cylinders to treat flux non-uniformity such as the resonance Doppler broadening in the actinide nuclides when temperature increases (Goltsev et al., 2003). In this context, the reactor codes that deal with such phenomena are very important tools because they make possible the calculations of neutron flux, criticality and burnup distribution in the nuclear fuel. These codes generally use stochastic methods such as Monte Carlo (Eckhardt, 1987) and deterministic methods like SDDM (Spatially Dependent Dancoff Method) (Matsumoto et al., 2005). In both cases it is necessary to validate the calculation methodology and their related nuclear data libraries by comparison with appropriate experimental data. The basic premise is how to apply microscopic cross sections in macroscopic systems of the reactor without losing the characteristics of the physical reality of the analyzed system.

Because of its importance in calculations, many of the existing nuclear data and a significant portion of all cross section processing codes are dedicated to treatment of the behavior of the actinide resonance self and mutual shielding (De Kruijf, 1993), especially in deterministic codes where homogenization of the materials present in the reactor core and reflector and multigroup treatment for the neutron energy domain are of major concern.

There has been a great interest in distribution measurements of nuclear reaction rates inside of the fuel rods and pellets (Andrianov et al., 2008; Matzke, 1992). This interest arose from the need to effectively understand the behavior of the resonance self and mutual shielding of the actinide nuclides such as ^{235}U, ^{238}U, ^{239}Pu and ^{240}Pu in the fuel pellets (Stoker and Weiss, 1996; Hwang and Wigner, 2004). Such measurements are used to homogenize the nuclear fuel burnup rate that varies along the radial direction of the fuel pellets optimizing core calculations.

* Corresponding author at: Instituto de Pesquisas Energéticas e Nucleares – IPEN/CNEN-SP, Centro de Engenharia Nuclear (CEN), Av. Prof. Lineu Prestes, 2242, 05508-000 – Cidade Universitária, São Paulo, SP, Brazil.
E-mail addresses: asantos@ipen.br (A. dos Santos), rjerez@ipen.br (R. Jerez).

http://dx.doi.org/10.1016/j.anucene.2016.09.046
0306-4549/

For several years, the mathematical methods and nuclear data libraries applied to the reactor physics have imposed severe restrictions on the calculated predictions. It was common to adopt empirical procedures such as cross sections adjustment to obtain better agreement between theory and experiment (Hellstrand, 1996). The main progresses in the comparison between theory and experiment are due to advances in the following points: improvement of nuclear data, new techniques of nuclear parameter measurements and a significant increase in computer processing speed. Nowadays, mathematical and numerical methods have matured to such a stage of development that it is believed that the major source of the uncertainty in the reactor physics codes reside in their basic nuclear data. Consequently, a major problem in the verification and validation of existing models to treat non-uniformity of nuclear transmutation is the lack of suitable experiments that provide possible physical quantities to be treated by these mathematical models. In this context the IPEN/MB-01 research reactor facility is extremely important. This facility consists of a 28×26 rectangular array of UO_2 fuel rods 4.3486 wt% enriched uranium as UO_2 and clad by stainless steel (SS-304) inside a light water tank. The maximum allowed power is 100 W. The control of the IPEN/MB-01 reactor is via two control banks diagonally placed. The square pitch of the IPEN/MB-01 reactor was chosen to be close to the optimum fuel-to-moderator ratio (maximum k_∞). This feature favors the thermal neutron energy region and mainly the ^{235}U fission events. This facility has well defined geometric and material data composition and many of its experiments have been accepted as international benchmarks in several critical configuration experiments (dos Santos et al., 2004) and in several other classical reactor physics experiments (dos Santos et al., 2005).

The distribution measurements of nuclear reaction rates along the radial direction of the fuel pellets are commonly performed using powerful techniques such as EPMA (electron probe microanalysis) and SIMS (secondary ion mass spectrometry). The spatial distribution of uranium, plutonium and certain fission products can be analyzed quantitatively through them. Both techniques have detection limits of approximately 10^{13} atoms per cubic centimeter. However, in the case of IPEN/MB-01 the fission rate is approximately 10^8 reactions/(cm^3-sec), so these techniques are not able to measure quantitatively the distribution of the secondary nuclear reaction products in the reactor due to its very low concentration.

In recent works carried out by Macku et al. (2006, 2007), direct and indirect measurements of nuclear reaction rates were carried out along the radial direction of fuel pellets using the SVEA-96 reactor Optima2, a BWR critical unit type in the PROTEUS complex (Williams et al., 1998). In the first work, direct measurements of capture reaction rate were made in the ^{238}U foils (C8) and fission measurements in ^{235}U foils (F5) that had been irradiated between the fuel rod pellets. After irradiation the foils were "cut" into rings and measured separately enabling sampling of four different radial regions of the fuel. The results of these measurements were compared to those calculated by MCNP-4C (Briesmeister, 2000) code and Casmo-4 code (Edenius et al., 1995). This type of measurement induced high uncertainty because part of the irradiated material is lost during the cutting process. In the second study, copper foils were used within the fuel rods for determining the spatial distribution of $^{63}CU(n,\gamma)^{64}Cu$ reaction. This reaction is of interest because the 1/v cross-section region of this reaction is similar to the ^{235}U fission reaction. The results were compared to those calculated with the MCNP-4C code. Although the shape of the cross section of $^{63}CU(n,\gamma)$ and $^{235}U(n,f)$ is similar below 0.3 eV, the radial fission profile was not fully determined.

The objective of this work is to present a new sampling technique based on the utilization of Pb collimators, making it possible to measure the radiative neutron capture in ^{238}U and the total fission rates in six different radial regions of an irradiated thin UO_2 disk employing the IPEN/MB-01 research reactor facility. The main advantage of this technique over the previous methods already presented is the maintenance of the integrity of irradiated disks during and after the measurements allowing the measurement to be repeated to verify its reproducibility. There is a great scarcity of experimental data of nuclear reaction rate distributions inside of the fuel pellets. This work aims to contribute to providing high quality experimental data, helping to address this deficiency and collaborating directly for code and related nuclear data library validations in the reactor physics area.

2. Experimental methodology and procedure

The nuclear reaction rates along the radius of the fuel pellets are obtained using the activation analysis technique employing a very thin UO_2 disk (about 0.9 mm thickness and 4.3% enrichment obtained from the UO_2 pellets of the facility). The gamma spectrometry of this disk is performed by the Gamma Detection System (GDS) which is composed of a very thin set of Pb collimators and a special HPGe (High Pure Germanium) detector. The Pb collimators are cylindrical and have concentric holes with variable diameters at their center. Because of its high sensitivity, activation analysis has been used in many IPEN/MB-01 experiments. The thermal flux in this facility is about 10^8 (n/cm^2 s) which is appropriate for many activation analysis experiments. This low neutron flux induces low radioactive levels in the irradiated targets, enabling their handling and reducing saturation effects in HPGe detectors.

The measurements of the radiative capture rate in ^{238}U and fission rates in both ^{238}U and ^{235}U require that the UO_2 disk be inserted between the fuel pellets of the fuel rod. An experimental demountable fuel rod, similar to the one used in the reactor was employed to cope with this task. Tests were conducted to demonstrate the UO_2 radial mass homogeneity in this fuel disk by measuring its thickness in several positions. That was a way to infer the mass homogeneity in the UO_2 disk. This UO_2 disk is inserted between the tenth and eleventh pellets of the rod; i.e. nearly 105.5 mm from the bottom of the active region of the core. This position is in the asymptotic region of the core and far enough from the control banks so that undesirable flux perturbation is prevented. The experimental fuel rod is irradiated at the central position of the core (position M-14 in Fig. 1) for 1 h at 100 W power level (maximum power of the IPEN/MB-01 reactor).

The core configuration in this experiment consists of an array of 28×26 with 680 fuel rods with 24 Ag–In–Cd control rods, which control the reactivity, and 24 B_4C safety rods kept at a position 19.20 cm above the active region of the core during the experiment. Fig. 1 shows the radial configuration in the experiments. A complete description of the IPEN/MB-01 reactor may be found in (Briggs, 2012).

Two irradiations, respectively, with and without cadmium sleeves are performed. In the first irradiation, a cadmium sleeve was wrapped around the experimental fuel rod at the disk position to discriminate thermal and epithermal neutrons from the second irradiation; i.e., the bare irradiation. The cadmium sleeve is 7.0 cm long and 0.5 mm thick and is inserted in the reactor core using the apparatus shown in Fig. 2. This apparatus has a Lucite ruler that is used to hold the cadmium sleeve. The height of the ruler can be adjusted relatively to the bottom of the fuel rod.

After irradiation, the UO_2 disk is taken to the Gamma Detection System (GDS), which performs the gamma spectrometry of the nuclear reaction products arising from the reaction rates occurring in ^{238}U and in ^{235}U. The UO_2 disk is covered with the Pb collimator so that the GDS system measures mainly the gammas emitted by