Transmutation rates determination for the study of nuclear fuel composition under irradiation

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ABSTRACT

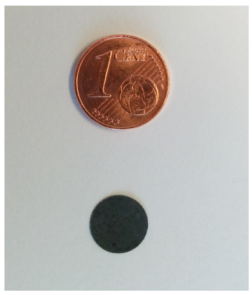
The prediction of the time evolution of the nuclear fuel composition is one of the key issues concerning the design and operation of nuclear plants. With this target in mind, a methodology, based on Monte Carlo codes calculations validated by direct measurements, was developed to estimate the transmutation rates of nuclides relevant for the study and analysis of nuclear fuel composition under irradiation, focusing on major and minor actinides build-up and burn-up and on fission product production and depletion.
Material targets containing the nuclides relevant for the study and analysis of nuclear fuel composition were irradiated in a well characterized irradiation position of the TRIGA Mark II reactor of Vienna Atominstitut in order to detect the transmutation rates (production & depletion) by means of gamma-ray spectrometry technique.
The experiment was then simulated using the Serpent-2 reactor model recently developed and validated as presented at the RRFM 2017 conference. Calculated and experimental results were then compared to verify the capability of Serpent-2 reactor model to predict changes in the composition of nuclear fuel under irradiation and are reported in this paper.

1. Introduction

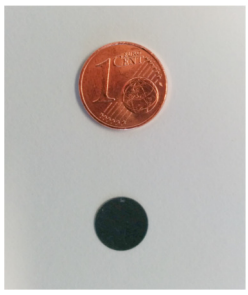
The TRIGA (Training Research and Isotope production General Atomics) MARK II reactor[1] at the Technische Universität Wien (TU Wien) is a pool-type research reactor moderated and cooled by light water, licensed for 250 kW steady state and up to 250 MW pulse operation. The core load during the activity described in this work consisted out of 76 stainless steel clad zirconium-hydride fuel elements (8.5%-wt enriched 19.95%-wt in U-235), in a cylindrical geometry.
Recent activity at the TRIGA reactor of TU Wien included the implementation and validation[2,3] of a new reactor model by means of the Monte Carlo code Serpent-2[4,5].
The present work describes another benchmark activity among Serpent-2 results and experimental data in an out-of-core irradiation position, for application in the field of transmutation rates determination. Selected material foils containing the nuclides relevant for the study and analysis of nuclear fuel composition were irradiated in the reflector annular groove irradiation facility (Lazy Susan) in order to detect the transmutation rates (production & depletion) by means of gamma-ray spectrometry technique. The experiment was then simulated using the Serpent-2 reactor model. Calculated and experimental results are here compared to verify the capability of Serpent-2 reactor model to predict changes in the composition of nuclear fuel under irradiation.

2. Experimental determination of transmutation rates

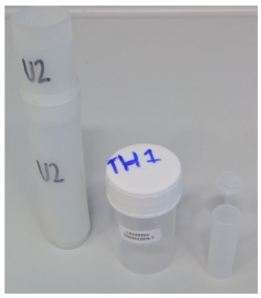
This activity was conducted in order to obtain experimental data about production and depletion of nuclides in natural Uranium (U) and Thorium (Th) foils following an irradiation at the TRIGA reactor of TU Wien.
Three samples of each kind (U, Th) were irradiated under consistent conditions in the Lazy Susan LS1 irradiation position. All samples were counted and spectra analyzed both before and after irradiation and the results compared.
The samples used for this irradiation at the TRIGA reactor were obtained by "Goodfellow Cambrige Limited" and were provided with appropriate certificates for the mass, dimensions and composition. In addition to the natural isotope composition of the Thorium and Uranium samples, some impurities are present in very low (ppm) concentrations but have no influence in the analysis.
The samples were thin foils with diameter of 10.0 mm. The Uranium foils present an average thickness of 0.178 mm, and the Thorium foils of 0.125mm.
From this point on the samples were assigned a code (U1, U2, U3 and TH1, TH2, TH3) used throughout irradiations and analysis process. U and Th samples in comparison to a cent coin are shown in Figure 1 together with the irradiation capsules utilized for the irradiation.



(a) Uranium foil compared with cent coin



(b) Thorium foil compared with cent coin.



(c) Irradiation Capsule.

Figure 1: Picture of Uranium (a), Thorium (b) foils and irradiation capsules (c).

Irradiation setting

Six irradiations were performed, one for each foil. The irradiation position was one of the dry beam tubes (LS1, Lazy Suzan 1) in the reflector annular groove irradiation facility. The irradiation time was 90 minutes, with reactor power of 5 kW. During each irradiation in LS1, a copper foil was irradiated in parallel in the Central Irradiation Channel (CIC) as a flux monitor, to allow eventual normalization of results.

Irradiated sample activity determination by gamma spectrometry

Irradiated foils activity were measured by means of a coaxial closed-ended HPGe n-type GAMMA-X with 52.8% relative efficiency, 1.81 keV energy resolution at 1.33 MeV and Peak/Compton edge ratio equal to 73.6. The efficiency calibration of the detector was performed by means of a certified solid multi-gamma calibration source (Type QCRB1186, Eckert&Ziegler) with dimension and geometry similar to those of the activated foils. The acquired spectra were analyzed using the GENIE2K[6] Analysis software provided from CANBERRA.
Since produced fission products present different half-lives, measurements were repeated for each foil at different elapsed times (≈10, 30, 100 days) after end of irradiation to better determine fission product activities. The best counting position (i.e. distance D between sample and detector) was selected over 3 possible calibrated

positions (D=180 mm, 250 mm, and 400 mm) finding a balance between low dead time and good counting statistics. The dead time was always kept under 10%.

Data analysis

The composition of the irradiated foils and the processes occurring during and after irradiation (neutron activation, decay, fission, etc.) were evaluated in advance to obtain an overview of the nuclides to check for in the gamma spectra. Peak interferences and mother-daughter pairs were also checked and corrected.

About U-235, when irradiated with thermal neutrons, a wide variety of fission products is produced. For determination of the fission fragments produced with the highest probability during the natural U foil irradiation, the fission products with Cumulative Fission Yield (CFY) >2.5% per fission of U-235 (Table 1) were selected for consideration according to the CFY values reported in literature[7].
Some of those nuclides (e.g. Ce-144, Sr-90) present a half-life too long for decent activity results in short spectroscopy measurements; others (e.g. I-135, Xe-135, Pr-144 and Nd-144) have instead a too short half-life for being still present at the time of measurement. In both cases then, those nuclides could not be detected.
For produced fission products that are in a parent-daughter relation (e.g. Te-132/I-132, Zr-95/Nb-95 and Ba-140/La-140) the activity of the daughter nuclide (d) is correlated with the decay of the parent nuclide (p). Hence, the daughter activity at end of irradiation ($A_d(0)$) was deducted by knowledge of the parent activity at end of irradiation $A_p(0)$ (derived by measured parent activity $A_p(t)$) and of the measured daughter activity $A_d(t)$, where t is the time of measurement.
In conclusion, nearly all fission products predicted were detected by means of gamma spectroscopy and the activation measurement results can be used for comparison with calculated results.

Element	CFY [% per fission]	Half Life		Element	CFY [% per fission]	Half Life
Zr-95	6.502	64.02d		Sr-90	5.73	28.81y
Nb-95	6.49	34.99d		Ru-103	3.10	39.27d
Mo-99	6.13	2.748d		Te-132	4.28	3.20d
(Tc-99m)	-	(6.01h)		I-131	2.88	8.02d
I-133	6.59	20.8h		I-135	6.39	6.57h
Xe-133	6.60	5.24d		Xe-135	6.61	9.14h
Cs-137	6.22	30.06y		Ba-140	6.31	12.77d
La-140	6.31	1.68d		Pr-144	5.47	17.28min
Ce-141	5.86	32.50d		Nd-144	5.47	-
Ce-144	5.47	285d		Nd-147	2.23	10.98d

Table 1: List of the U-235 fission products that where taken into account for the analysis, their Cumulative Fission Yield (CFY) and half-lives (Tc-99m was expected as a product of Mo-99 decay).

For the irradiated Thorium foils, among the spectroscopy measurement performed the ones at about 30 days after irradiation resulted as the more appropriate for the analysis.
The Thorium samples present a natural abundance of 100% Th-232. During the irradiation, fissile Th-233 is produced by capture reaction of thermal neutrons. Th-233 then decays according to the decay chain shown in Figure 2, with production of fissile U-233. Due to the short half-life of Th-233, Pa-233 was the only detectable nuclide from this activation decay chain.
Among the fission product of U-233 only Ba-140 and La-140, having the highest cumulative fission yield of 6.24, were produced in very slow amount and detected.

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow[22.15min]{\beta^-} {}^{233}Pa \xrightarrow[26.98d]{\beta^-} {}^{233}U$$

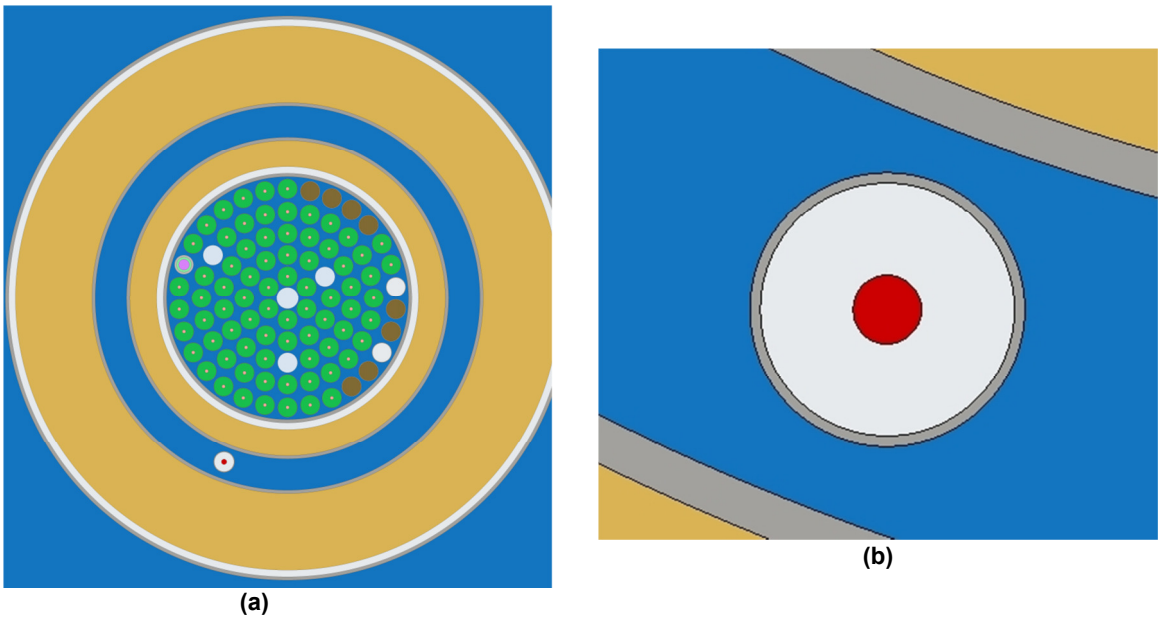
$$^{232}Th \xrightarrow[1.41E10]{\alpha} {}^{228}Ra \xrightarrow[5.7y]{\beta^-} {}^{228}Ac \xrightarrow[6.1min]{\beta^-} {}^{228}Th \xrightarrow[1.9y]{\alpha} {}^{224}Ra$$

$$^{224}Ra \xrightarrow[3.6d]{\alpha} {}^{220}Rn \xrightarrow[55s]{\alpha} {}^{216}Po \xrightarrow[0.14s]{\alpha} {}^{212}Pb \xrightarrow[10.6h]{\alpha} {}^{212}Bi$$

Figure 2: The capture reaction on Th-232 with its decay chain (up to U-233 production) and the natural Th-232 decay chain are shown.

3. Experiment simulation with Serpent

The three-dimensional model of the TRIGA reactor in Vienna was developed with Serpent-2 and verified[2,3] in the recent period.
This model contains, at the proper level of detail, all essential core components that can affect the evaluation of the neutron flux in-core distribution and the energy spectrum in different in-core positions, including inside the fuel elements; as well as the reactor fuel burn-up and reactor critical parameters. The Serpent-2 reactor model extends up to the reactor tank, including the graphite reflector, the horizontal beam tubes and the annular groove irradiation facility (Lazy Suzan).
The reactor horizontal and vertical sections of Serpent-2 model are shown in Figure 3(a) and 4(a) respectively. In both figures, it can be seen the annular groove irradiation facility (Lazy Suzan) that penetrates the graphite reflector and is filled with water. In the Lazy Susan, five aluminum vertical dry irradiation tubes are installed for irradiation of small samples: the one (named LS1) used for the present experiment is reproduced in Serpent and is visible in detail in Figures 3(b) and 4(b). The natural U/Th foil in the irradiation position is represented in red color.



(a) (b)

Figure 3: (a) The horizontal section of the TRIGA reactor model as obtained with Serpent-2 and (b) the detail of irradiation facility LS1 used for U and Th foils irradiation.

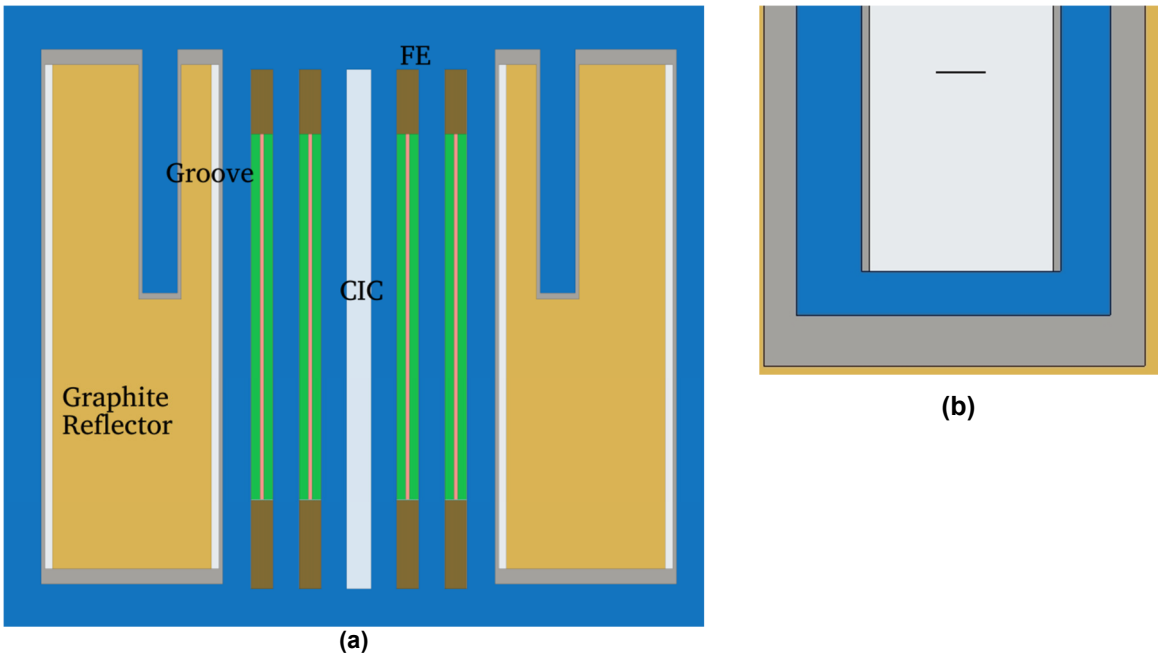


Figure 4: (a)The vertical section of the TRIGA core modeled with Serpent-2. (b) detail of irradiated foils in the LS1 irradiation position.

The simulation options were 1 million source neutrons per cycles, with in total 1500 cycles. To simulate the irradiation, the burn up of the foils material was modelled by Serpent with burn up time of 90 minutes and 5 kW reactor power.
As Serpent can calculate the activities of every possible generated nuclide, the output requirement included all the nuclides measured in the experiment (see Table 1).
Calculated results are compared with experimental values in Figure 5 for U foils and in Figure 6 for Th foils.

In case of the Uranium foils, most determined activities are in a good agreement (below 10%) between Serpent and the experiment; Serpent results are comparable with the measurement results within the uncertainties. However, it can be observed that some isotopes have bigger discrepancy between simulation and experimental data. This is the case of Tc-99m, Ce-141, Te-132 and Xe-133: the explanation could be that the gamma lines for detection of those isotopes lay in the low energy region (below 230 keV), where the uncertainty of the detector is very large.
In case of the Thorium foils, the Serpent simulation matches the experiment within the uncertainty (8%) except for Pa-233, where the discrepancy rises to about 27%.

When comparing the results of the simulation with experimental data, one must be aware that the simulation can have some inaccuracies. The neutron flux spectrum in out of core region could for example not results exactly the same as in the reality. Additionally, Serpent burn calculation in thin and small volumes (like the irradiated foils) is not accurate as in larger volumes (like for example the fuel elements).

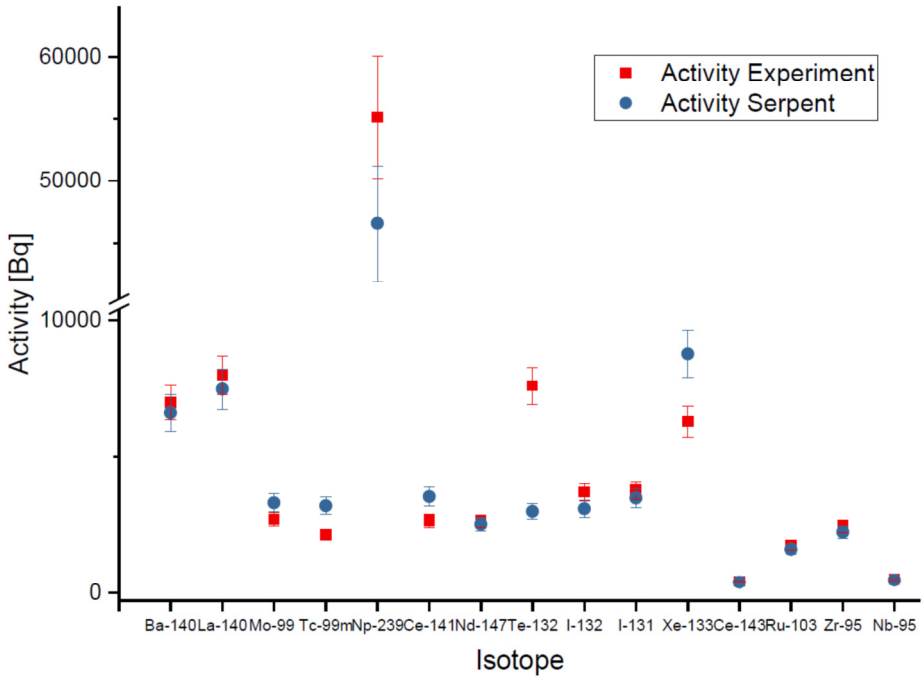


Figure 5: Calculated and experimental activity values of the Uranium-foil.

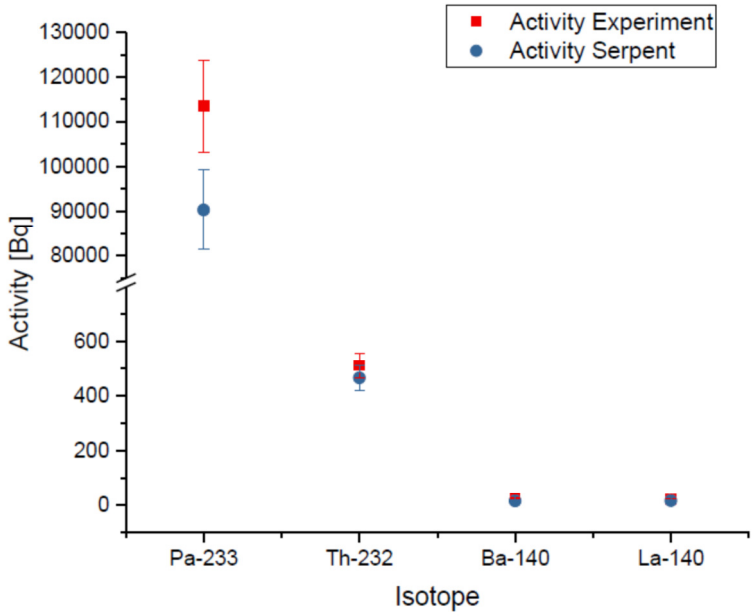


Figure 6 Calculated and experimental activity values of the Thorium-foil.

4. Discussion and conclusions

This work was intended for the study of a methodology, based on Monte Carlo codes calculations validated by direct measurements, to estimate the behaviour of nuclear fuel composition under irradiation, focusing on major and minor actinides build-up and burn-up and on fission product production and depletion.

Material foils containing nuclides (U and Th) relevant for the study and analysis of nuclear fuel composition were irradiated in an out-of-core irradiation position of the

TRIGA Mark II reactor of the TU Wien. Transmutation rates were then detected by means of gamma-ray spectrometry technique.
The experiment was then simulated using the Serpent-2 reactor model. Calculated and experimental results were compared to verify the capability of Serpent-2 reactor model to predict changes in the composition of nuclear fuel under irradiation.
In conclusion, the comparison shows that calculated Serpent-2 results are in a good agreement with the experimental ones within the uncertainties of the measurement. However, in few cases a bigger discrepancy is observed between simulation and experimental data. This happens for some fission products detected trough gamma lines in the low energy region (<230 keV), where the detector is less accurate and the uncertainty larger.
Other factors in the simulation also carry some inaccuracies that can affect the results. For example, the region of interest in this work is an out-of-core region: it has to be considered that the neutron flux spectrum in this region could not exactly reproduce the real flux distribution. Additionally, Serpent burn calculation in thin and small volumes is not accurate as in larger volumes: i.e. the calculation in the irradiated foils volume hardly would provide same accuracy as a burn-up calculation in a fuel element volume located in the core.

5. References

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