2017 International Nuclear Atlantic Conference - INAC 2017
Belo Horizonte, MG, Brazil, October 22-27, 2017
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR – ABEN

Agência Brasileira do ISBN
ISBN 978-85-99141-07-6

ANALYSIS OF BURNUP OF ANGRA 2 PWR NUCLEAR WITH ADDITION OF THORIUM DIOXIDE FUEL USING ORIGEN-ARP

Isadora C. Gonçalves[1], Caio C. Wichrowski[2], Claudio L. de Oliveira[3], Sergio O. Vellozo[4], and Camila O. Baptista[5]

[1] Instituto Militar de Engenharia - Seção de Engenharia Nuclear
Praça Gen Tibúrcio, 80
22290-270 Rio de Janeiro, RJ
isadora.goncalves@ime.eb.br

[2] Instituto Militar de Engenharia - Seção de Engenharia Nuclear
Praça Gen Tibúrcio, 80
22290-270 Rio de Janeiro, RJ
wichrowski@ime.eb.br

[3] Instituto Militar de Engenharia - Seção de Engenharia Nuclear
Praça Gen Tibúrcio, 80
22290-270 Rio de Janeiro, RJ
d7luiz@yahoo.com.br

[4] Instituto Militar de Engenharia - Seção de Engenharia Nuclear
Praça Gen Tibúrcio, 80
22290-270 Rio de Janeiro, RJ
vellozo@ime.eb.br

[5] Instituto Militar de Engenharia - Seção de Engenharia Nuclear
Praça Gen Tibúrcio, 80
22290-270 Rio de Janeiro, RJ
camila.oliv.baptista@gmail.com

ABSTRACT

It is known that isotope ^{232}thorium is a fertile nuclide with the ability to convert into ^{233}uranium, a potentially fissile isotope, after absorbing a neutron. As there is a large stock of available thorium in the world, this element shows great promise in mitigate the world energy crisis, more particularly in the problem of uranium scarcity, besides being an alternative nuclear fuel for those currently used in reactors, and yet presenting advantages as an option for the non-proliferation movement, among others. In this study, the analysis of the remaining nuclides of burnup was carried out for the core configuration of a PWR (pressurized water reactor) reactor, specifically the ANGRA II reactor, using only uranium dioxide, its current configuration, and in different configurations including a mixed oxide of uranium and thorium in three concentrations, allowing a preliminary assessment of the feasibility of the modification of the fuel, the resulting production of ^{233}uranium, the emergence of ^{231}protactinium (an isotope that only occurs as a fission product of ^{232}Th) resulting from burning. The study was carried out using data obtained from FSAR (Final Safety Analysis Report) of ANGRA II, using the SCALE 6.1, a modeling and simulation nuclear code, especially its ORIGEN-ARP module, which analyzes the depletion of isotopes presents in a reactor.

1. INTRODUCTION

Thorium has been researched and utilized, experimentally, in the initial stages of nuclear reactors development. Due to the necessity of appropriate material for military artifacts, political issues and sovereignty of technology, through the decades, uranium has become the preferential choice as primary fuel for the nuclear reactors. The possible scarcity of uranium among other reasons, such as non-proliferation policies, has opened a way back for the thorium to be the object of research and development again. As it is known, Brazil have one of the largest thorium reserves in the world and its utilization may become of great relevance to the country [01]. Therefore the study of remaining nuclides of the burnup considering Angra II PWR at its actual configuration has been developed, first using uranium dioxide as a fuel, and, then using a mix oxide of thorium and uranium, allowing a prior evaluation of the fuel depletion for this modification and the consequent production of ^{233}uranium, the emergence of protactinium and resultant poisons of the burnup, as well some advantages of this new fuel insertion.

2. FUNDAMENTATION

Thorium is a chemical element, more specifically a radioactive actinide metal, and was discovered in 1829 by Jöns Berzelius. In 1898 its radioactive characteristics were discovered by Marie Curie and Gerhard Carl. This element is generally found in nature with almost total purity of its isotope ^{232}Th [01], which is fertile (process shown in Figure 1) and capable to induce breeding, it means that during its fertilization process, which is characterized by the absorption of a neutron and transmutation into a fissile element, the number of neutrons emitted by the fissile isotope resultant (in this case ^{233}U) is bigger than the number of neutrons spent in the process [02].

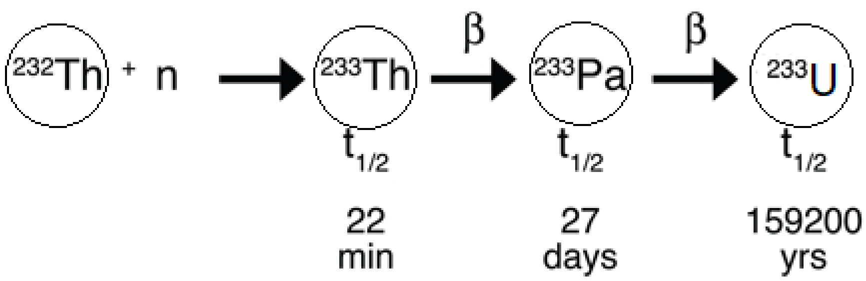

Figure 1 - ^{232}Th fertilization process and transmutation into ^{233}U [02].

Due to this characteristic, thorium can be used as a nuclear material [03], more specifically as a reactor fuel, in several energy ranges, mainly in thermal and epithermal reactors due to its neutron yield rate per absorbed neutron in reactors of these types as demonstrated in Figure 2 [04], as it is an option for the most commonly used fuels as well as in the form of a tool for the non-proliferation process, since when associated with plutonium, thorium, in its fertilization, is able to "spend" the plutonium itself and consequently its military potential [05].

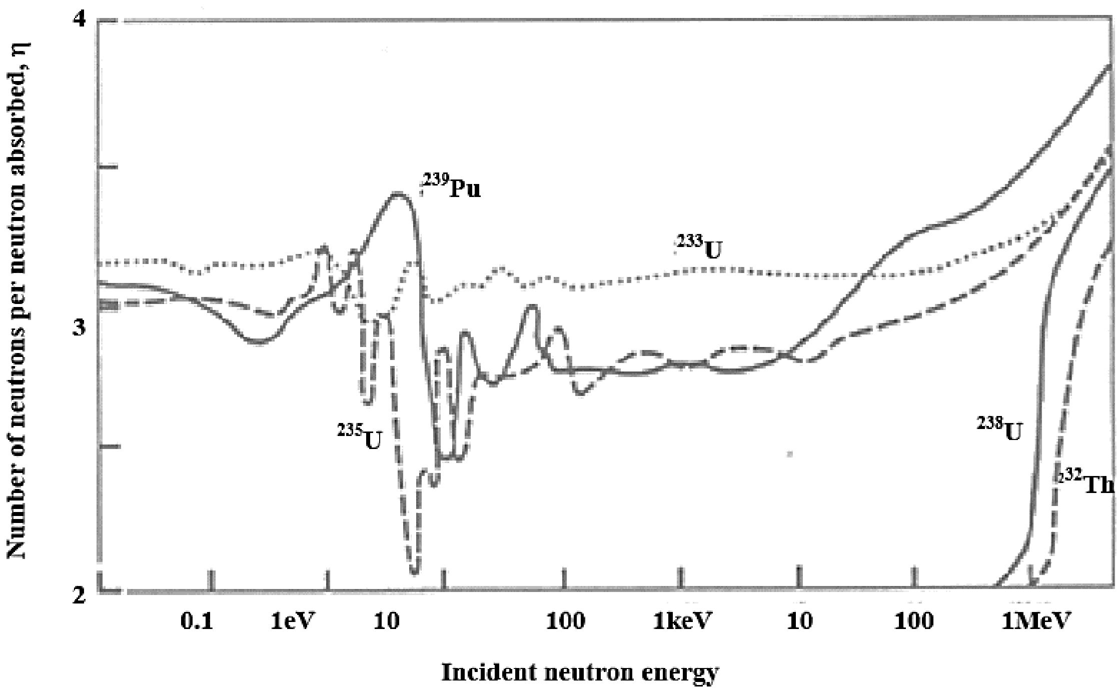

Figure 2 - Neutron yield per neutron absorbed [04].

Thorium presents several advantages in its use, because it is three to four time more abundant than uranium [01], have more stable chemical characteristics, support higher temperatures of fuel burnup, as well the ThO_2 have 10% more thermal conductivity than UO_2 [05]. In addition, the separation of fissile isotopes is harder at fuels based on thorium, what would make it difficult at the process of proliferation [05]. Then, in Table 1, some nuclear data of the main fissile and fertile isotopes for the thermal neutron range are presented.

Table 1 – Neutronic properties of some isotopes in the thermal zone [06].

Nuclear Data		^{232}Th	^{233}U	^{235}U	^{238}U	^{239}Pu	^{241}Pu
Cross section (barns)	Fission	0.000052	529.10	582.6	0.000003	748.1	1011.1
	Capture	7.35	45.50	98.8	2.68	269.3	362.1
Resonance (barns)	Fission	0.00	775.00	275.0	0.00163	303	570
	Capture	83.3	138.00	146.0	277.00	180	192
υ		0.00	2.48	2.43	0.00	2.87	2.97
η_{th}		0.00	2.26	2.08	0.00	1.91	2.23

3. METHODS

In this analysis it was performed a simulation of the PWR Angra II reactor using the SCALE 6.1, a nuclear code of modeling and simulation, developed by the Reactor Nuclear Systems Division (RNSD) of the Oak Ridge laboratory, more specifically the included ORIGEN-ARP (Oak Ridge Isotope Generation – Automatic Rapid Processing) module, that analyze depletion, decay and radioactive source for the nuclides generated during the operation of a nuclear reactor, the burnup. The analysis was made in two steps, first the actual configuration had been analyzed, contending only ^{238}U with an enrichment of 3.2% of ^{235}U. After that, other analysis, with different configurations of thorium dioxide addiction, had been made.

3.1. Origen-Arp

The ORIGEN-ARP, is a nuclear code simulates the depletion/decay conditions for fuels based on uranium alternating cross session libraries of ORIGEN with the enrichment, fuel burnup and the reactors moderator density. Also, is an analysis module from SCALE, this sequence uses the ORIGEN-S code, independent cross session libraries and interpolations of the algorithms that operate according to the fuel proprieties and operating conditions [07].

3.2. Data Analysis

All the information for this work were taken from the Angra II FSAR. The Angra II reactor is a German designed and Brazilian built PWR which has thermal capacity of up to 3782 MWt generating 1350MWe. Light water acts as moderator and cooling agent and must operate under high pressure to keep the coolant as a liquid.

Each fuel assembly is arranged in a matrix of 16x16 rods placed in a square cross section with symmetrical quarters. There are 5 control rods and 59 fuel rods per quarter. The fuel assemblies are placed in an almost cylindrical configuration with different enrichment regions. The initial loading has fuel assemblies filled with uranium dioxide pellets with three different enrichment levels: 1.9%, 2.5% and 3.2%. Each fuel rod is made of pellets stacked inside a Zircaloy 4 cylinder (cladding) and the void between them is filled with helium [08].

For the simulation of burnup, as already mentioned before, it was chosen a PWR with Siemens German technology, the Angra II, operating with uranium dioxide with 3.2% enriched fuel, density of 10.4 g/cm^3 and a total of 623 tons of mass [08]. The irradiation situation with 0,4949772 MTU to a burnup of 30MW on a period of 90 days was chosen, looking to the generation of actinides.

Three reactor configurations were chosen, the first one with the core entirely fueled with uranium dioxide 3.2 % enriched, the second fueled with 90% of uranium dioxide 3.2 % enriched and 10% of thorium dioxide, and the third situation fueled with 80% of uranium dioxide 3.2 % enriched and 20% of thorium dioxide all the mixtures in a mass basis. The value utilized of thorium dioxide density for the simulation was 10g/cm^3, taken from the Compendium [09].

4. RESULTS

After being collected the data related to the PWR Angra II and being realized the simulations of burnup using the ORIGEN-ARP module, the calculated quantities of burnup nuclides were determined and are presented on the Table 2.

Table 2 - Quantities (atoms/barn.cm) of isotopes generated by the burnup of Angra II with different concentrations of ^{232}Th.

Quantity of ^{232}Th added (%)	Isotopes	Time (days)			
		0	30	60	90
0%	^{231}Pa	0,00E+00	5,43E-14	1,05E-13	1,50E-13
	^{239}Pu	0,00E+00	1,79E-05	3,45E-05	4,81E-05
	^{241}Pu	0,00E+00	5,88E-08	4,00E-07	1,16E-06
	^{232}Th	0,00E+00	1,02E-14	3,98E-14	8,72E-14
	^{233}U	0,00E+00	1,41E-12	2,64E-12	3,70E-12
	^{235}U	7,52E-04	7,07E-04	6,64E-04	6,25E-04
	^{236}U	0,00E+00	8,30E-06	1,60E-05	2,31E-05
	^{238}U	2,25E-02	2,24E-02	2,24E-02	2,24E-02
10%	^{231}Pa	0,00E+00	1,28E-08	2,52E-08	3,66E-08
	^{239}Pu	0,00E+00	1,76E-05	3,36E-05	4,63E-05
	^{241}Pu	0,00E+00	6,91E-08	4,59E-07	1,30E-06
	^{232}Th	2,28E-03	2,27E-03	2,26E-03	2,26E-03
	^{233}U	0,00E+00	2,49E-06	7,72E-06	1,38E-05
	^{235}U	6,77E-04	6,32E-04	5,91E-04	5,53E-04
	^{236}U	0,00E+00	8,20E-06	1,57E-05	2,25E-05
	^{238}U	2,02E-02	2,02E-02	2,02E-02	2,01E-02
20%	^{231}Pa	0,00E+00	2,84E-08	5,56E-08	8,03E-08
	^{239}Pu	0,00E+00	1,73E-05	3,25E-05	4,42E-05
	^{241}Pu	0,00E+00	8,21E-08	5,29E-07	1,45E-06
	^{232}Th	4,56E-03	4,54E-03	4,52E-03	4,51E-03
	^{233}U	0,00E+00	5,52E-06	1,71E-05	3,03E-05
	^{235}U	6,01E-04	5,57E-04	5,18E-04	4,81E-04
	^{236}U	0,00E+00	8,08E-06	1,53E-05	2,18E-05
	^{238}U	1,80E-02	1,79E-02	1,79E-02	1,79E-02

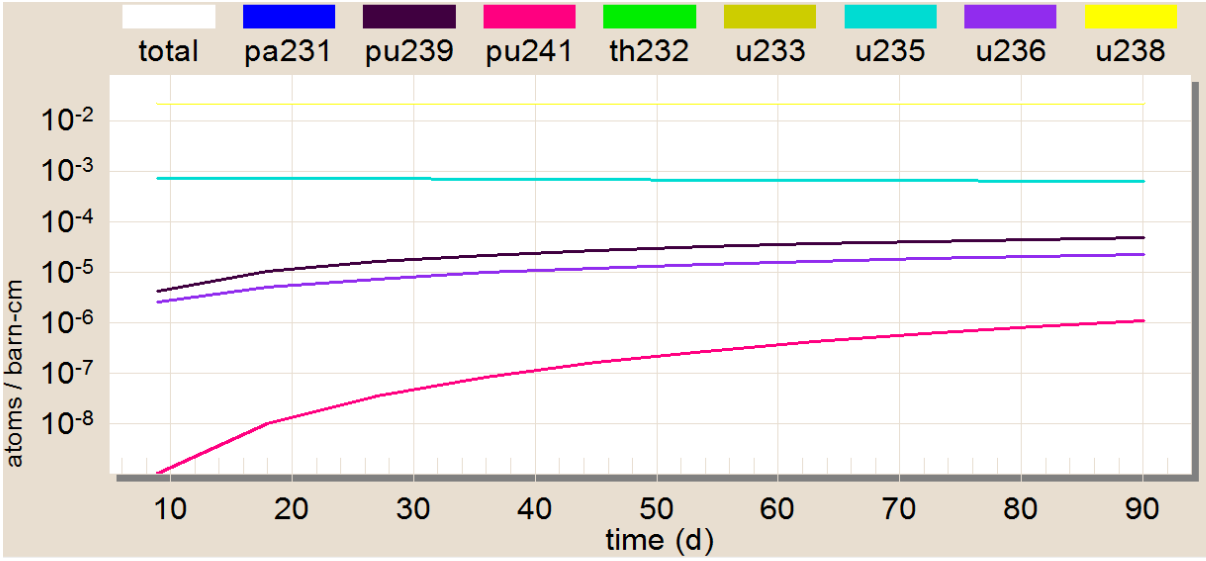

Figure 3 - Isotopes generated by the burnup of Angra II with 0% of ThO_2.

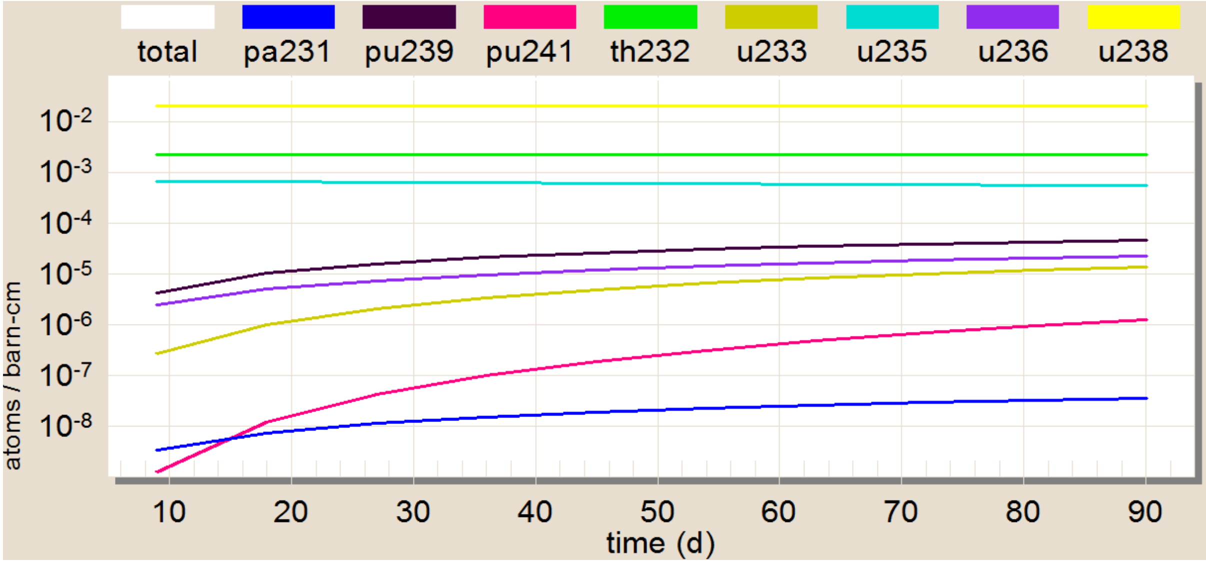

Figure 4 - Isotopes generated by the burnup of Angra II with 10% of ThO_2.

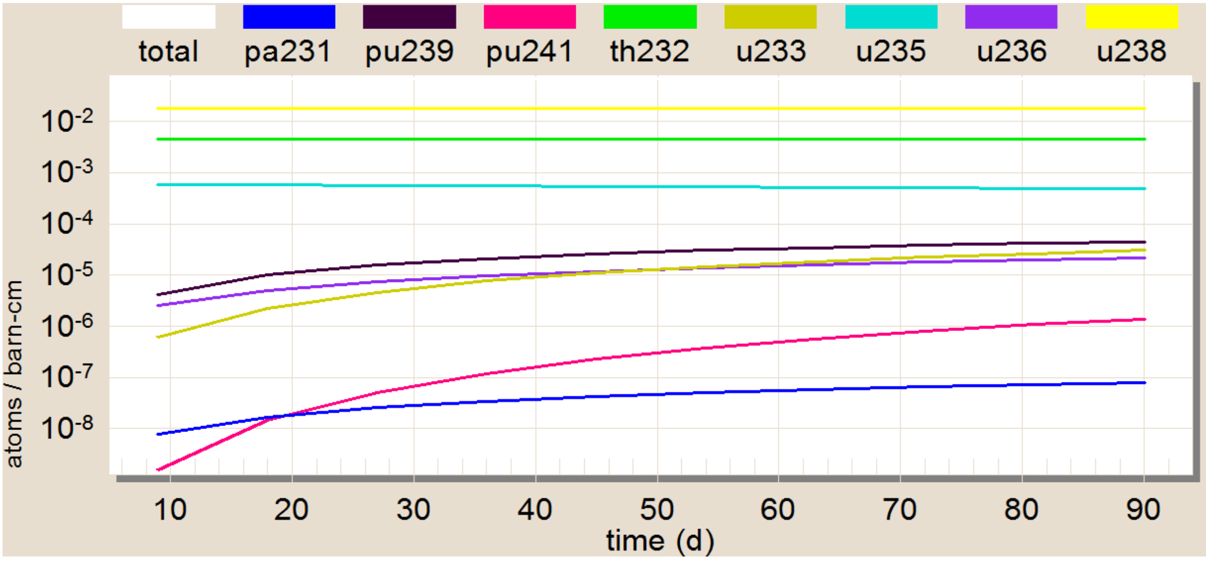

Figure 5 - Isotopes generated by the burnup of Angra II with 20% of ThO_2.

After analyzing the data resulting from the simulation, it was realized that when 10% of thorium dioxide was added a change on the behavior of the generated isotopes of the burnup has occurred such that, the quantity of ^{231}Pa has been almost duplicated, with proves that ^{232}Th is reacting, since the ^{231}Pa only occurs as a fission product of ^{232}Th [10]. The ^{233}U quantity almost triplicate from the 30th to the 60th day, that is the main objective of the insertion of ^{232}Th, although both are very small when compared to the ^{232}Th quantity does not show a measurable change, as expected, since most of the neutrons are absorbed in the uranium yet it is possible to observe the generation of fissile material. It should be noted that some of the ^{233}U had already ben fissioned.

Observing the generation data and graphs, another thing that was also be realized, is that the ^{238}U fertilization is bigger than the ^{232}Th, because even if the microscopic cross sessions capture of ^{232}Th is three time higher than the ^{238}U (Table 1), the macroscopic cross sessions capture of ^{238}U get still bigger, due to its higher quantity into the reactor.

.

5. CONCLUSIONS

Considering only the materials, when analyzing the nuclear data of ^{232}Th one realizes that its capture cross session of neutrons in thermal range (7.35 barns) is almost three times higher than the ^{238}U (2.68 barns), another fertile nuclide, but capture resonance of ^{232}Th (83.3 barns) is lower than the ^{238}U (277 barns) what means that thorium based reactors need a “starting” isotope with a higher enrichment, what shouldn’t be a real problem to Brazil, that have an enrichment technology by centrifuges very well developed. Even if the microscopic cross session capture of ^{235}U and ^{239}Pu, and the υ be higher than ^{233}U, even so there is an advantage on the use of ^{233}U, because its fission resonance and mainly its exploitation rate (ηth) is the best considering those nuclides, in other words, ^{233}U spend less neutrons than the other fissile isotopes.

As higher the insertion of thorium dioxide on the reactor and more time of burnup, higher is the production of ^{233}U, and it is believed that the number of generations isn’t higher just due

to the macroscopic cross capture session of ^{232}Th being low, because on all simulated situations the quantity of thorium was always much lower than the ^{238}U total.
In general, the burnup nuclides of ^{233}U and of ^{235}U are equal, and with very similar concentrations. One of few isotopes that appear with more frequency on the ^{233}U fission and not on the ^{235}U is the ^{231}Pa, that in real in a product of the ^{232}Th fission.
It may be concluded that the thorium is a great fuel, with a lot of advantages, and its use is viable on PWR, as Angra II, together with the uranium dioxide, as far as at the beginning of its cycle there is a greater "feeding" of neutrons or a decrease at the controlling neutrons absorbing isotopes of the reactor, due to in this way there is continuity on the fertilization process.
To the full analysis of the PWR Angra II behavior with the addiction of thorium dioxide and verification of the viability is still necessary to perform realize some studies, that may be developed further, as simulations of the reactors core to study the resultant poisons of the fission burnup, the verification of criticality of the system as the thorium is added, the evaluation of the physical-chemical aspects of the mix oxide of thorium and uranium fuel, thermodynamic and hydraulic behavior of the PWR Angra II reactor with the addiction of the thorium dioxide, among other things.

REFERENCES

1. J. L. Robert, *Mineralogy of Uranium and Thorium*, Schiffer, Pennsylvania, United Statesof America (2016).
2. Belle, J.; Berman, R.M. *Thorium Dioxide properties and nuclear applications*, Office of Naval Reactors, Washington, United States of America (1984).
3. IAEA, Safeguards Glossary. International Nuclear Verification Series n°3 (2001).
4. IAEA, *Thorium fuel cycle — Potential benefits and challenges*. Nuclear Fuel Cycle and Materials Section International Atomic Energy Agency, Vienna, Austria (2005).
5. M. Yousif Alhaj, Nader M.A. Mohamed, Alya Badawi, Hanaa H.Abou-Gabal, "Towards proliferation-resistant thorium fuels," *Annals of Nuclear Energy*, V. 101, March 2017, Pages 586-590 (2017).
6. "Live Chart of Nuclides," https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html (2017).
7. ORNL. *Scale: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design.* Oak Ridge : Oak Ridge National Laboratory (2011).
8. Eletronuclear, Eletrobrás Termonuclear S.A. "*Final Safety Analysis Report - FSAR: Central Nuclear Almirante Álvaro Alberto, Unit 2*", Rio de Janeiro, Brazil (1999).
9. RJ McConn Jr, *Compendium of Material Composition Data for Radiation Transport Modeling, Department of Energy Contract*, United States of America (2011).
10. "WIMS Library Update Project," https://www-nds.iaea.org/wimsd/fpyield.htm (2017).