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Reduction of Plutonium Production in Heavy Water Research Reactor: A Feasibility Study through Neutronic Analysis Using MCNPX2.6 and CINDER90 Codes

H. Shamoradifar, B. Teimuri, P. Parvaresh, S. Mohammadi

***Abstract*—**One of the main characteristics of Heavy Water Moderated Reactors is their high production of plutonium. This article demonstrates the possibility of reduction of plutonium and other actinides in Heavy Water Research Reactor. Among the many ways for reducing plutonium production in a heavy water reactor, in this research, changing the fuel from natural Uranium fuel to Thorium-Uranium mixed fuel was focused. The main fissile nucleus in Thorium-Uranium fuels is U-233 which would be produced after neutron absorption by Th-232, so the Thorium-Uranium fuels have some known advantages compared to the Uranium fuels. Due to this fact, four Thorium-Uranium fuels with different compositions ratios were chosen in our simulations; a) 10% UO_2-90% THO_2 (enriched= 20%); b) 15% UO_2-85% THO_2 (enriched= 10%); c) 30% UO_2-70% THO_2 (enriched= 5%); d) 35% UO_2-65% THO_2 (enriched= 3.7%). The natural Uranium Oxide (UO_2) is considered as the reference fuel, in other words all of the calculated data are compared with the related data from Uranium fuel. Neutronic parameters were calculated and used as the comparison parameters. All calculations were performed by Monte Carol (MCNPX2.6) steady state reaction rate calculation linked to a deterministic depletion calculation (CINDER90). The obtained computational data showed that Thorium-Uranium fuels with four different fissile compositions ratios can satisfy the safety and operating requirements for Heavy Water Research Reactor. Furthermore, Thorium-Uranium fuels have a very good proliferation resistance and consume less fissile material than uranium fuels at the same reactor operation time. Using mixed Thorium-Uranium fuels reduced the long-lived α emitter, high radiotoxic wastes and the radio toxicity level of spent fuel.

***Keywords*—**Burn-up, heavy water reactor, minor actinides, Monte Carlo, proliferation resistance.

I. Introduction

HEAVY water research reactors are typically fueled with natural uranium, which contains about 99.3% of the isotope uranium-238 and about 0.7% of the fissile isotope uranium-235. Reactors fueled with natural uranium are well suited for plutonium production. Plutonium is produced when a U-238 nucleus absorbs a neutron and then undergoes two consecutive beta decays. Given the high percentage of U-238 in natural uranium, the plutonium production rates in heavy-water reactors are significantly higher than other reactors [4], [8]. There are many ways to reduce plutonium production and MA inventory in a heavy water research reactor. One of them is to load Thorium-based fuels in such reactors. Thorium fuels have a chemical stability, better thermal conductivity, and great abundance. These fuels create a low quantity of MA, and high quality of waste, so thorium has been researched as a nuclear fuel in the countries with nuclear power industry such as United States, United Kingdom, Germany, and some other countries that have a high amounts of thorium fuel sources [2], [3], [7].

In this work, thorium-uranium as fuel in reactor core was considered. MCNPX2.6 code is used to calculate neutronic parameters while CINDER90 code is also used for Burn-up calculations.

II. Methodology

Four types of Thorium-Uranium fuels with different compositions in our simulations have been used and have been loaded separately in the core to calculate neutronic parameters such as Burn-up, MA inventory, fissile isotopes production and reactor poisons concentration variations, axial and radial neutron fluxes. All of the simulations were performed by MCNPX2.6 code and also Burn-up has been calculated by CINDER90 code. KCODE with 7000 initial neutrons, 250 effective cycles and 50 ineffective cycles have been applied for calculations of neutronic parameters. In order to validate the code, some HWZPR (Heavy Water Zero Power Reactor) experiments were simulated by MCNPX2.6 code, and the calculated results were compared with experimental data.

A. Monte Carlo Method

The purpose of all transport calculations, in general, is the determination of the particle flux $\varphi(\vec{r}, E, \vec{\Omega})$ and the functions of it, such as reaction rates, doses, currents, etc. [1]. The flux is often obtained as the solution of the Boltzmann transport equation and as such is grasped as a deterministic quantity because it is a solution of a deterministic equation related to the exact quantities. Monte Carlo methods are very different from deterministic transport methods [3]. By contrast, Monte Carlo does not solve an explicit equation, but rather obtains answer by simulating individual particles and recording some

H. Shamoradifar is with the Department of Nuclear Physics, Payame Noor University (PNU), P.O.Box 19395-3697, Tehran, Iran (Corresponding author; e-mail: H.Shamoradifar@gmail.com)
B. Teimuri is with the Nuclear Science and Technology Research Institute, Atomic Energy Organization of Iran.
P. Parvaresh and S. Mohammadi are with the Department of Nuclear Physics, PNU, P.O.Box19395-3697, Tehran, Iran.

aspect (tallies) of their average behavior. The average behavior of particles in the physical system is then inferred from the average behavior of the simulated particles [3]. Therefore, one can say that Monte Carlo solves a transport problem by simulating particle histories rather than by solving an equation. Since the Monte Carlo method involves the generation of a large number of particle histories, using a considerable amount of nuclear data, its use is very strongly dependent on computer time and memory.

B. Introduction to MCNPX and CINDER90 Codes

The Monte Carlo N-Particle transport code system (MCNPX) developed by Los Alamos National Laboratory (LANL), is a general-purpose code for calculating the time-dependent continuous energy transport for neutrons, photons, electrons, or coupled Neutron/photon/electron in three-dimensional geometry [5]. The code includes the capability of calculating eigenvalues for critical systems. For neutrons, energy region is from 10^{-11} to 20 MeV, and all reactions given in a particular cross-section evaluation (such as ENDF/B-VII) are accounted for thermal neutrons are described by both free gas and S (α,β) models. MCNPX2.6 code is used for many applications such as, Burn-up, nuclear criticality safety, radiation shielding, nuclear safeguards, detector design and analysis, accelerator target design, medical physics and radiotherapy including BNTC, PET and neutron and photon oncology.

The Burn-up calculation involves a MCNPX2.6 calculations linked to a deterministic depletion calculations. CINDER90 code does the depletion calculations to calculate the nuclide densities, as a function of time after decaying. This code has inherent decay and 63-group cross section data library for 3400 isotopes [5]. MCNPX2.6 code then takes those new number densities and generates another set of fluxes and reaction rate; the process repeats itself until after the final time step [6].

C. Heavy Water Research Reactor Description and Simulation

Simulated system is a tank-type reactor with pressure tubes, heavy-water coolant and moderator, and 19-pin fuel bundles in a hexagonal lattice. The reactor is postulated to fuel with natural uranium. Its experimental facilities include a vertical thimble along the central axial line of the tank (Central Test Loop), one vertical 15-centimeter-diameter medical beam tube, and three vertical 10-centimeter-diameter research beam tubes. By these facilities, doing some activities such as fuel testing, radioisotope production, neutron radiography, neutron depth profiling (NDP), neutron activation analysis (NAA), and many other experiments are possible. It assumes that the pressure tubes and fuel's cover are made of Zircaloy-2. Dimensions of the center tube are the same in the inside and outside and are filled with coolant. The schematic model of reactor, as simulated by MCNPX2.6, is depicted in Figs. 1 and 2.

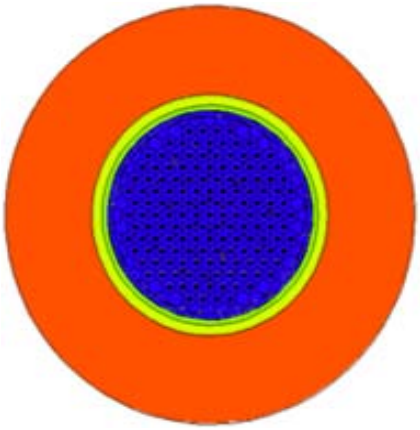

Fig. 1 Reactor core layout simulated by MCNPX (Z=0)

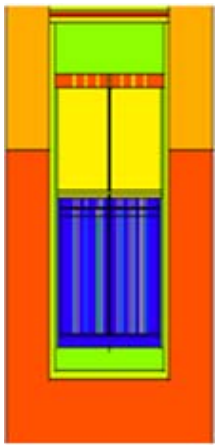

Fig. 2 Schematic model of reactor as simulated by MCNPX (Y=0)

D. Fuel Compositions / The Right k_{eff}

After benchmarking and reactor simulating, the next step was to find the correct fuel compositions. The correct fuel enrichment and weight percent of UO_2 was found by varying the weight percent of UO_2 in the THO_2-UO_2 mixture from 4% to 35%, with different degrees of enrichment from 3.7% to 20%. In the first step for each case, theoretical fuel density and neutronic parameters such as k_{eff} (multiplication factor) were calculated and compared with the related data from Uranium fuel. So base on obtained results it was decided to focus on four different fissile fuel components that are presented in Table I.

TABLE I
FUEL COMPOSITION

	Fuel	Enrich (%)	Density(g/cc)	k_{eff}
A	10%UO_2-90%THO_2	20	10.3171	1.13728
B	15%UO_2-85%THO_2	10	10.3176	1.02604
C	30%UO_2-70%THO_2	5	10.3189	1.06480
D	35%UO_2-65%THO_2	3.7	10.3194	1.01754
E	UO_2	Natural	10.4	1.05231

III. Result and Discussion

In this study, with burn up calculations, the potential of reducing the plutonium production and MA inventory of thorium-uranium fuels for heavy water research reactor has been investigated. According to computational data, the obtained burn-up results suggest some changes on the nuclides mass and compositions of spent fuels. The mass balance of plutonium isotope and MA inventory for thorium-uranium fuels and also neutronic parameters (k_{eff}, flux…) were compared with the same parameters of UO_2 fuel.

A. Burn-Up Calculations

1. Evolution of Multiplication Factor and Neutron Flux

The evaluated effective multiplication factor (k_{eff}) for four considered thorium-uranium fuels and reference UO_2 fuel is shown in Fig. 3. The results indicate that k_{eff} for all of fuels decreases near the zero time due to ^{135}Xe build up and then decrease again smoothly due to the fuel burn-up. For 10% UO_2-90% THO_2 (enriched=20%) fuel, the multiplication factor has a lowest and for natural UO_2 fuel, it has highest descending slope during burn-up time.

Neuron flux variations over burn-up time are shown in Fig. 4. The results indicate that the neutron flux increase with burn-uptime which is due to the consumption of fissile isotopes and increasing of the number of fissions. Also the neutron flux values and variations for all of fuels are almost similar (10^{13}n/s•cm^2), it is therefore perfectly suited for the purposes of research. Fuels (15% UO_2-85% ThO_2) and (35% UO_2-65% THO_2) have maximum and minimum neutron flux respectively.

2. Mass Changes of Fissile Isotopes

Mass changes of fissile isotopes ^{235}U, ^{233}U and ^{239}Pu as a function of burn-up time are shown in Figs. 5-8.These figures show that the main fissile nuclide^{235}U is consumed, while fissile nuclides ^{233}U and ^{239}Pu are produced.

Fig. 5 shows mass changes of ^{235}U isotope as a function of burn-up time. It is clear that after 360 days, the amount of ^{235}U for 10% UO_2-90% THO_2 fuel has the maximum value (163.2 kg) and the minimum consume (10.4%), while the amount of ^{235}U isotope for natural UO_2 has the minimum value (70.2 kg) and the maximum consume (21%). Hence, the utilization efficiency for ^{235}U in UO_2 fuel is higher than others.

The ^{233}U which has a very small cross section for neutron capture, produced from thorium and often occurs in most fissions. ^{233}U is the best fissile nucleus because it has the smallest ratio of capture to fission cross section, as low as 0.11. The production of fissile ^{233}U during the burn-up time can be seen in Fig. 6, but it does not increase linearly, because the fission rate increases with the increase of production. The amount of ^{233}U for 15% UO_2-85% THO_2 fuel is higher than the other fuels.

As we know ^{239}Pu is formed through the capture of a neutron in^{238}U and two consecutive β decays of^{239}U. The mass changes of ^{239}Pu as a function of burn-up and burn-up time are presented in Figs. 7 and 8, respectively. It is obvious in presented figures that the amount of ^{239}Pu mass for natural uranium fuel is very higher than Thorium-Uranium based fuels, because of the higher percentage of ^{238}U (99.2%) than the other fuels that can produce large amounts of ^{239}Pu. The amount of produced ^{239}Pu mass for UO_2 fuel (11970 gr) in 450 days is about 15.9 times more than 10% UO_2-90% THO_2 fuel (748.7gr), 8.4 times more than 15% UO_2-85% THO_2 fuel (1422 gr), almost 4.7 times more than 30% UO_2-70% THO_2 fuel (2531 gr) and about 3.64 times more than 35% UO_2-65% THO_2 fuel (3288 gr). Also it is clear from Fig. 7 that that fuel burn-up for Thorium-Uranium mixed fuels is higher than UO_2 fuel.

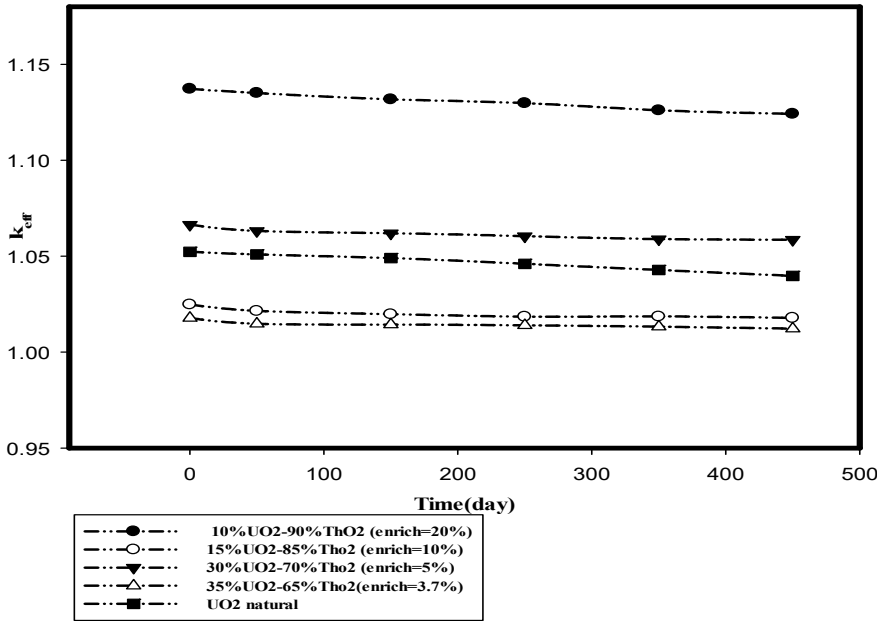

Fig. 3 Multiplication factor (k_{eff}) as a function of burn-up time

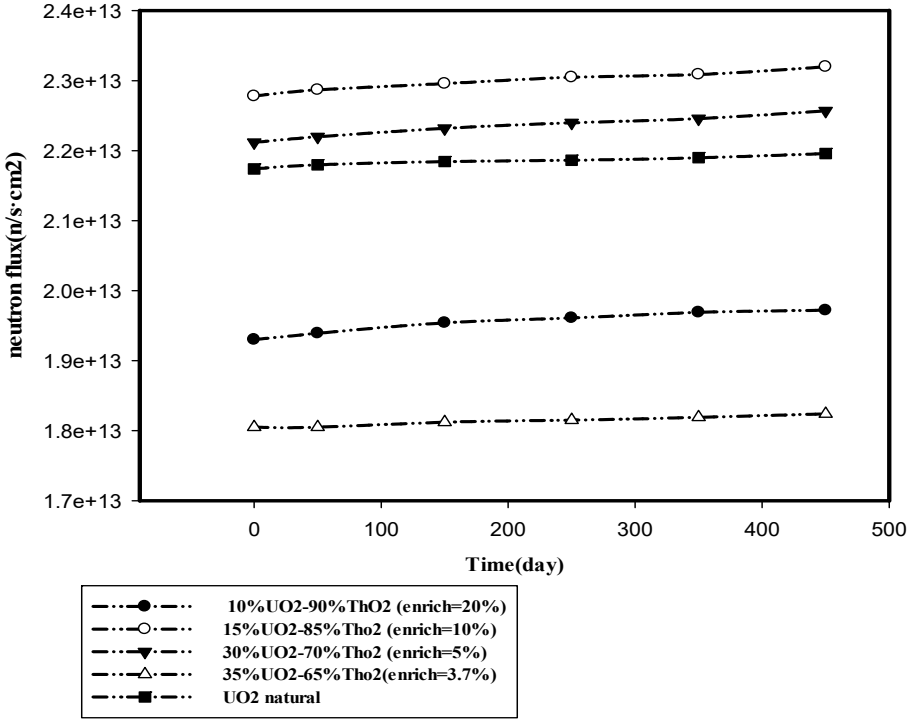

Fig. 4 Neutron flux as a function of burn-up time for the different fuel loads

Fig. 9 presents the fuel burn-up as a function of time for Thorium-Uranium and Uranium fuels. The results show that the fuel burn-up for thorium-uranium fuels decreased with increasing of the uranium percentage in the fuels; therefore, it is obvious that 10% UO_2-90% THO_2 fuel has the maximum burn-up, and natural UO_2 fuel has the minimum burn-up.

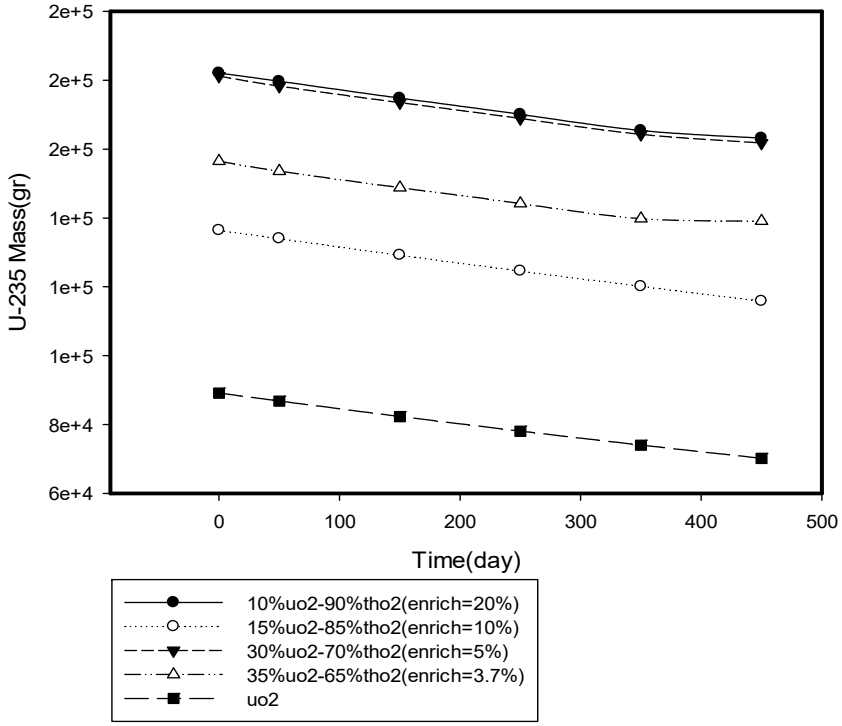

Fig. 5 Variation in the mass of U-235 isotope as a function of Burn-up time

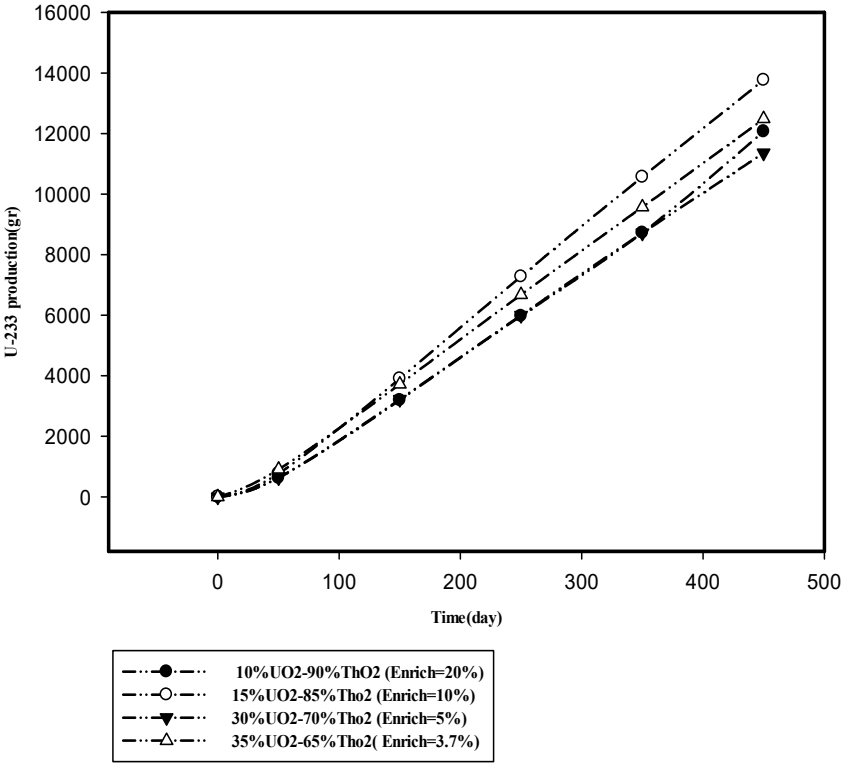

Fig. 6 Variation in the mass of U-233 isotope as a function of Burn-up time

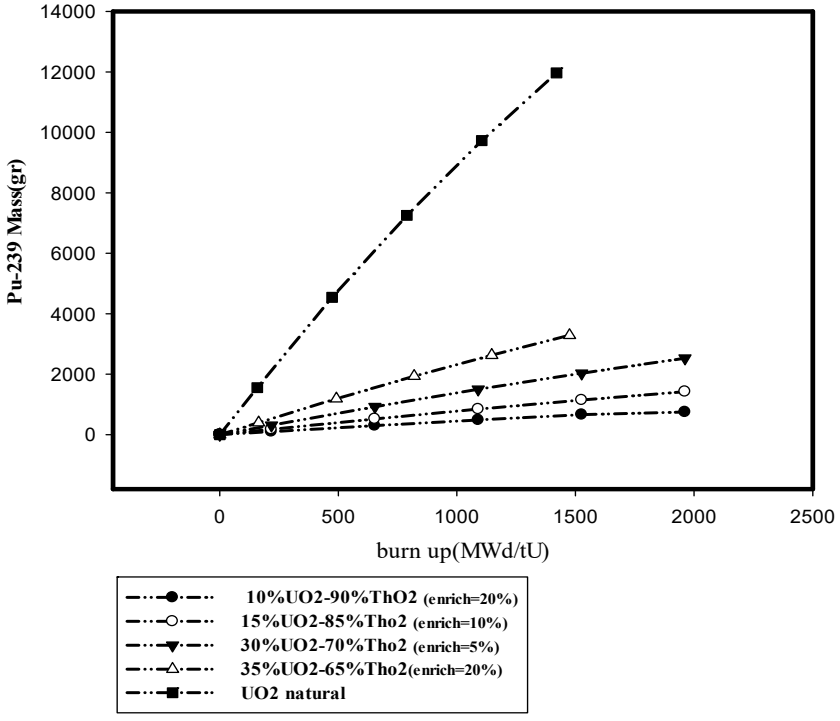

Fig. 7 Variation in the mass of PU-239 isotope as a function of Burn-up time

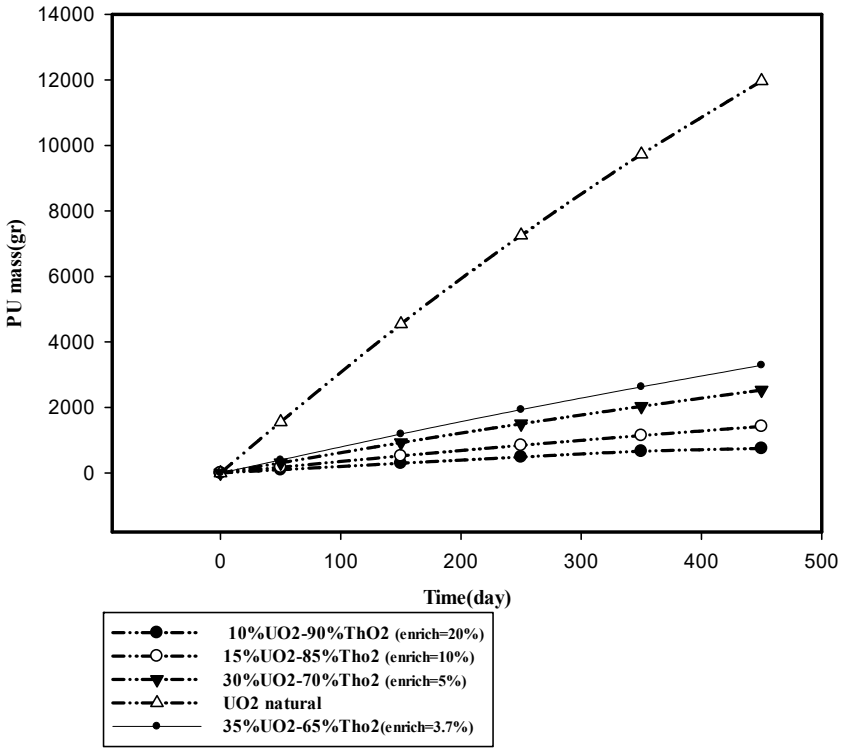

Fig. 8 Variation in the mass of PU-239 isotope as a function of Burn-up time

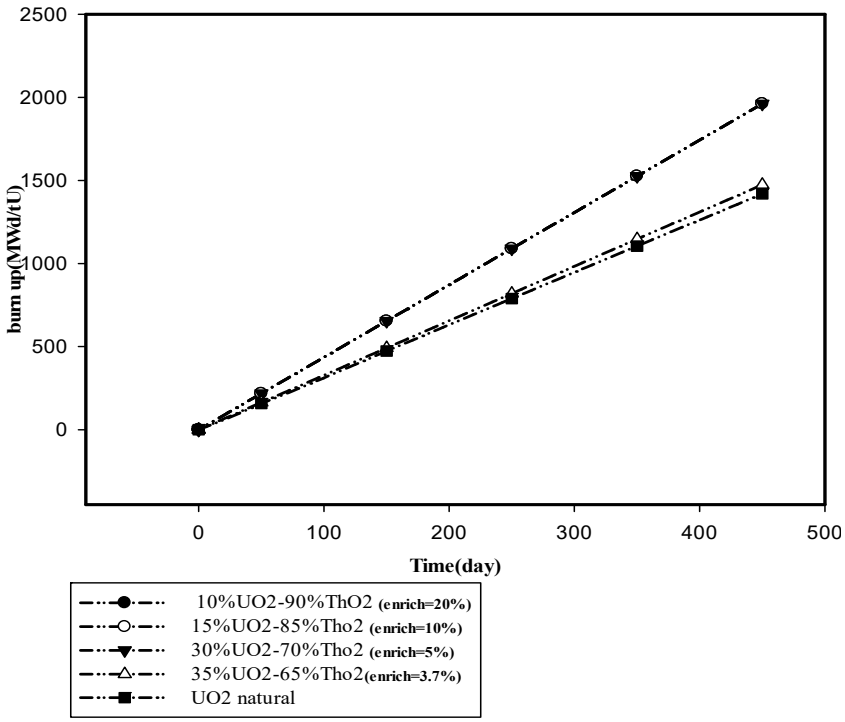

Fig. 9 Burn-up behavior as a function of time

3. Inventory of MA

Long-term potential radio toxicity of spent fuel arises principally from the presence of transuranics actinides (Pu and the so-called MA Np, Am, Cm, etc.) that have long half-lives and strong α – decaying and is produced based on following reactions:

$$^{241}_{95}Am \xrightarrow{(n,\gamma)} {}^{242}_{95}Am \left(\xrightarrow{\beta^-(16.02\ h)} {}^{242}_{96}Cm\right)$$

$$^{239}_{94}Pu \xrightarrow{(n,\gamma)} {}^{240}_{94}Pu \xrightarrow{(n,\gamma)} {}^{241}_{94}Pu \xrightarrow{\beta^-(14.35\ yr)} {}^{241}_{95}Am \left(\xrightarrow{\alpha(432.2\ yr)} {}^{237}_{93}Np\right)$$

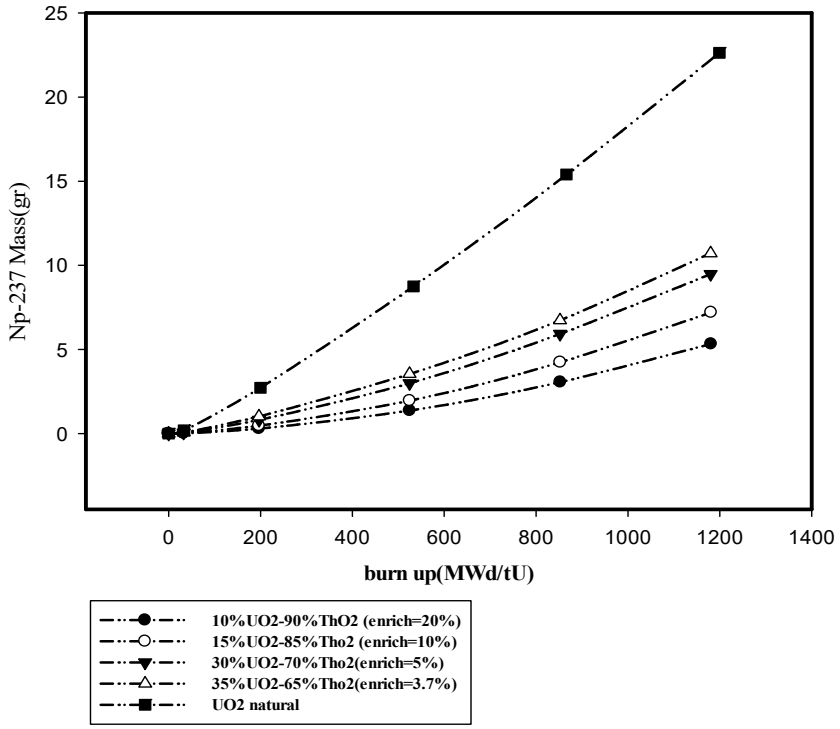

Fig. 10 ^{237}Np mass variations for the all of fuels

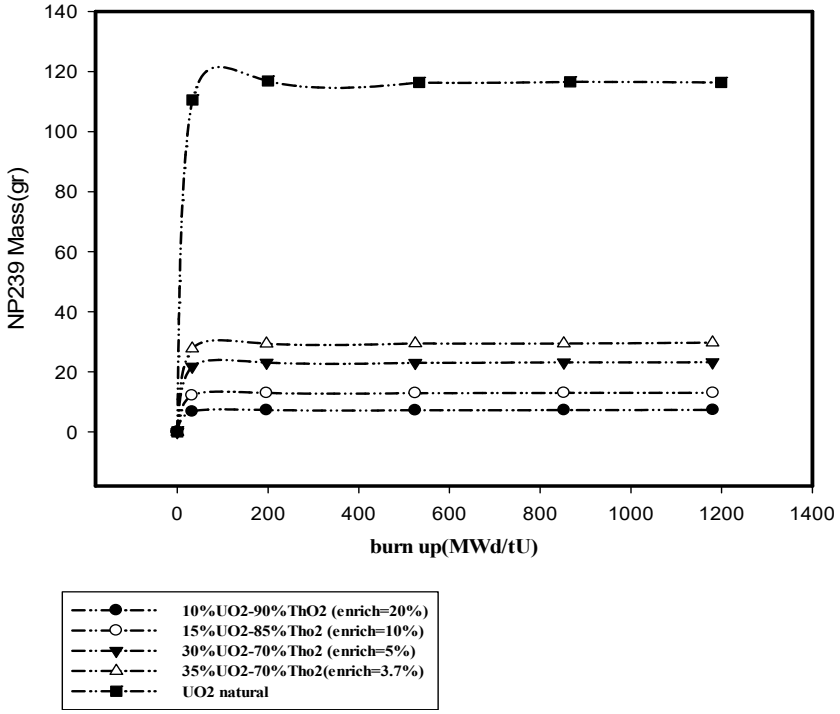

Fig. 11 ^{239}Np mass variations for the all of fuels

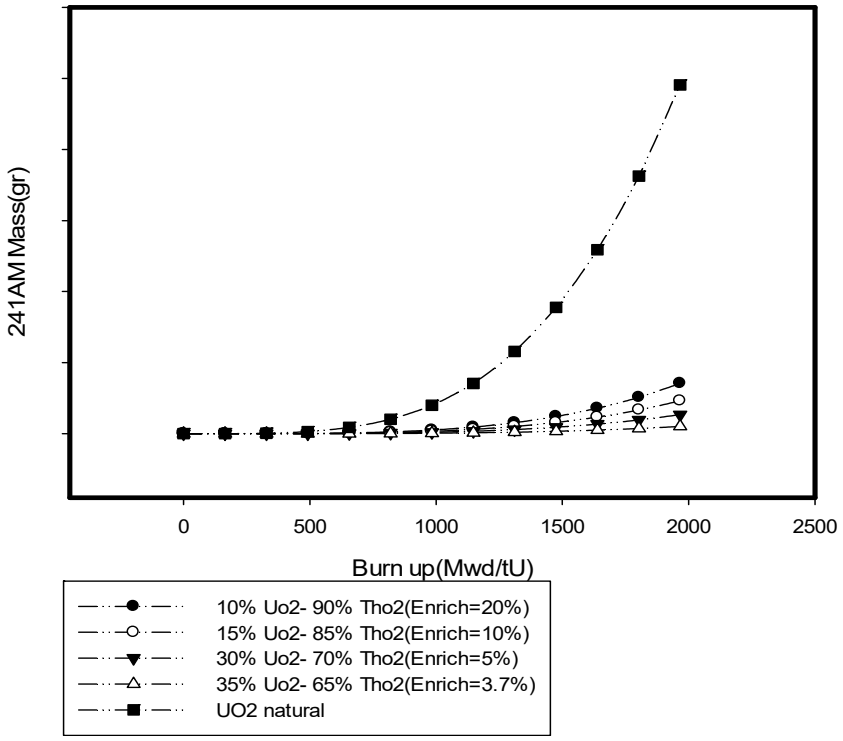

Fig. 12 ^{241}Am mass variations for the all of fuels

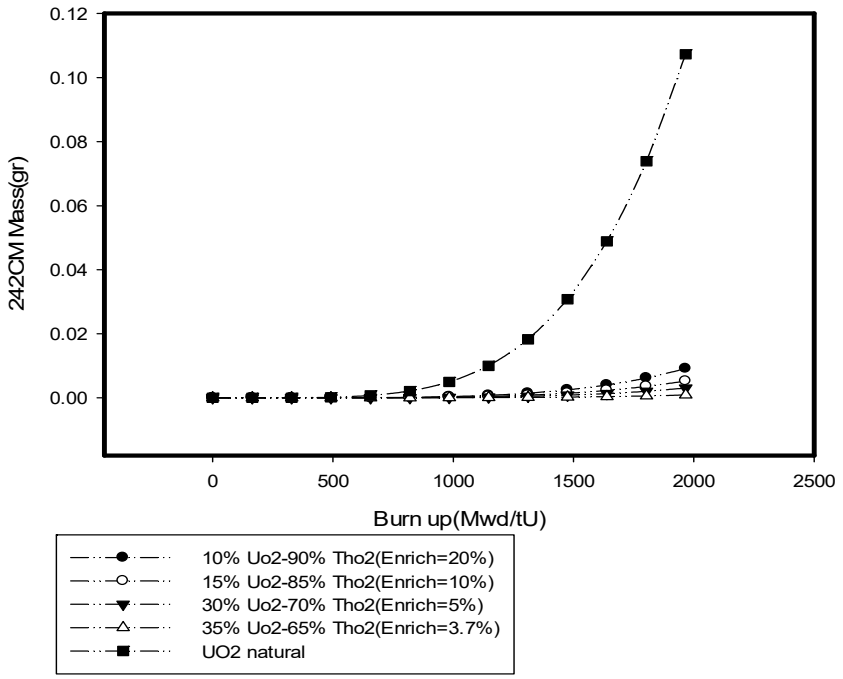

Fig. 13 ^{242}Cm mass variations for the all of fuels

Mass variations of the MA as a function of burn-up for Thorium-Uranium fuel and natural uranium fuel in a reactor are presented in Figs. 10-13. Based on these figures, it can be concluded that the production of MA for the (Thorium-Uranium) fuels is extremely low in comparison with natural uranium fuel, and also show the probable benefits of implementation of the Th fuel cycle.

4. Fission Products as Reactor Poison (^{135}Xe and ^{149}Sm)

The next step in this research was to find the amount of

neutron poisons such as ^{135}Xe and ^{149}Sm that have a big neutron capture cross section and have impact on the thermal utilization factor and thus multiplication factor and reactivity fluctuations of a reactor.

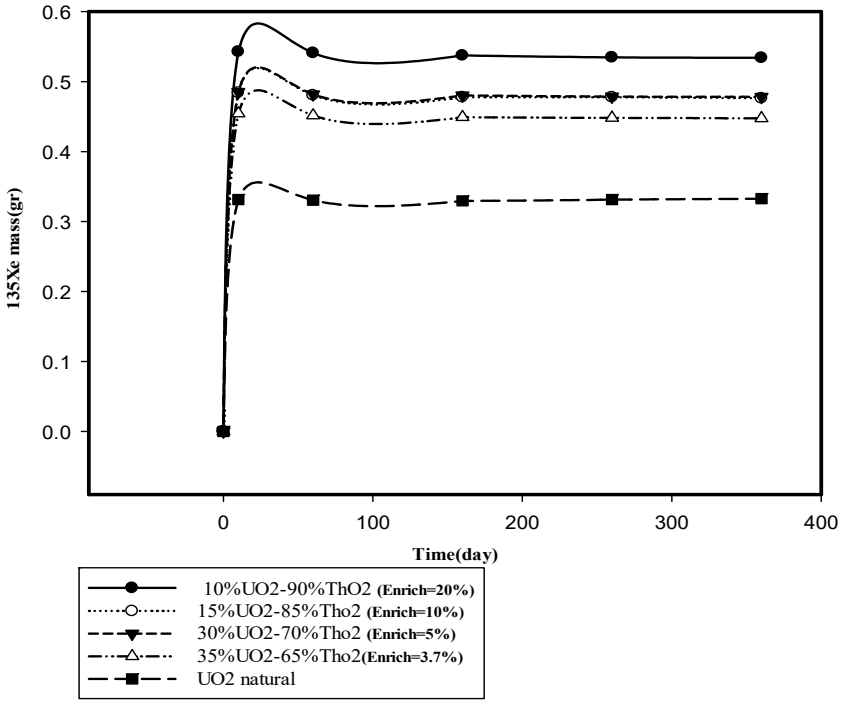

Fig. 14 ^{135}Xe concentrations as a function of Burn-up time

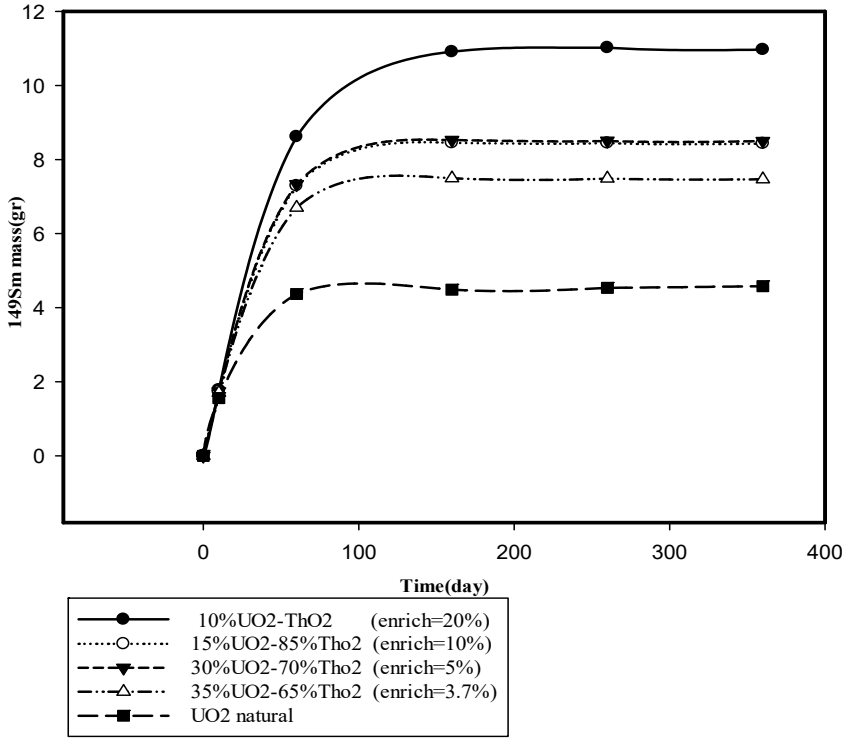

Fig. 15 ^{149}Sm concentrations as a function of Burn-up time

Fig. 14 shows Xenon-135 build-up versus fuel burn-up time; the results show that the concentration of Xenon for all fuels, in near zero time, is maximum, which is causing sharp decline in the amount of k_{eff} at the beginning point. Also a build-up of Xenon-135 in Thorium-Uranium fuel is more than Uranium fuel.

The ^{149}Sm mass production versus fuel burn-up time for all of fuels has been presented in Fig. 15. As the figure shows, during the Thorium-Uranium fuels burn-up ^{149}Sm has the highest production value, while the ^{149}Sm mass production for UO_2 fuel is lowest.

IV. Conclusion

According to the obtained results of burn-up calculations for Thorium-Uranium fuels and natural Uranium fuel, it is obvious that Thorium-Uranium can be used as a fuel in a heavy water research reactor. These kinds of fuels have several advantages and some disadvantages related to the natural Uranium. Thorium-Uranium fuels could dramatically reduce plutonium production, long-lived α emitter isotopes (MA), high radiotoxic wastes production and also cause to lower descending of k_{eff} and reactivity on time. For Thorium-Uranium, the fuel burn-up is decreased with increasing of uranium percent in the fuel. The 10% UO_2-90% THO_2 fuel has a maximum burn-up, and the natural UO_2 has a minimum burn-up, so high burn-up in Thorium-Uranium fuels are considered as an advantage over UO_2 fuel. Based on calculated results Thorium based fuels have higher value and lower consume of ^{235}U than natural uranium fuel, therefore the utilization efficiency of ^{235}U is higher for UO_2 fuel. Radial and axial flux changes process for all of fuels are almost the same for all of the considered fuels. Neutron poisons produced during reactor operation will also higher with Thorium-Uranium fuels than natural uranium fuel and this disadvantage of Thorium-Uranium fuels should be considered in the reactor design with these kinds of fuels.

References

[1] Thorium. (2006). Retrieved August 31, 2006 from http://www.world-nuclear.org/info/inf62.htm
[2] M.S. Kazimi, "Thorium fuel for nuclear energy," American Scientist, 91(5), 408, Retrieved August 1, 2006.
[3] M.S. Kazimi, R.K. Czerwinski, M.J. Driscoll, P. Hejzlar, J.E. Meyer, "On the use of thorium in light water reactors," (Academic Report no. MIT-NFC-TR-016). Cambridge, Massachusetts: Massachusetts Institute of Technology, August 1, 2006.
[4] A. Ahmad, F.V. Hippel and A. Glaser, "Conversion Options for Iran's IR-40 Reactor with Reduced Plutonium Production," Program on Science and Global Security, Princeton University, 2014.
[5] G. W. McKinney, J. W. Durkee, J. S. Elson, M. L. Fensin, J. S. Hendricks, M. R. James, R. C. Johns, D. B. Pelowitz, L S Waters T A Wilcox L. S. Waters, T. A. Wilcox, "MCNPX 2. 7 0 – New Features Demonstrated," Nuclear Engineering and Nonproliferation Division, LANL IEEE NSS/MIC Anaheim, CA, October 29 - November 3, 2012.
[6] J. S. Hendricks, G.W. McKinney, M.L. Fensin, M. R. James, R.C. Johns, J. W. Durkee, J. P. Finch, D. B. Pelowitz, L. S. Waters, M. W. Johnson, "MCNPX 2.6.0 Extensions," Los Alamos National Laboratory, LA-UR-08-2216, APRIL 11, 2008.
[7] R.S. Kemp, "Two Methods for Converting Iran's IR-40 Reactor to Use Low-Enriched-Uranium Fuel to Improve Proliferation Resistance after Startup," Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, March 2014.
[8] N. Prasad, A. Kumar, U. Kannan, A. Kumar, P.D. Krishnani and R.K. Sinha, "Study for use of LEU along with Thorium in Advanced Heavy Water Reactor (AHWR) to Enhance Proliferation Resistance Characteristics of Fuel," Bhabha Atomic Research Centre2015.