Neutronic evaluation of thorium-uranium fuel in heavy water research reactor

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ABSTRACT *High production of plutonium is the main feature of Heavy Water Moderated Reactors. However, reducing plutonium and other actinides in heavy water research reactors is possible. Changing the required fuel from natural uranium to the mixed thorium-uranium fuel was considered for reducing the production of plutonium in heavy water reactors. Thorium-uranium fuel is somewhat advantageous to uranium fuel. Thus, for the purpose of our study a simulation was done and four different types of thorium-uranium fuels with different composition ratios were selected for that. Natural uranium oxide was regarded as the reference fuel. Neutronic parameters were considered as our parameters for the required comparisons. Monte Carlo N-Particle eXended (MCNPX2.6) steady state reaction calculation linked to a deterministic depletion calculation (CINDER90) was used to do the calculations. The obtained results indicated that thorium-uranium fuel has appropriate resistance to proliferation as compared to uranium fuel. Long-lived α, radio toxicity level of used fuel, and wastes with high radio toxicity are reduced by applying the mixed thorium-uranium fuels.*

KEYWORDS*: Burn-up, heavy water reactor, minor actinides, Monte Carlo, proliferation resistance.*

I. Introduction

Heavy water research reactors mainly use natural uranium fuel with 99.3% of uranium isotope-238 (238U) and 0.7% of fissile uranium isotope-235 (235U). These reactors are appropriate for producing plutonium, which is made through absorption of neutron by 238U and is affected afterwards by two consecutive beta decays. Heavy water reactors produce plutonium more than other types of reactors, which is due to high rate of uranium isotope (238U) in natural uranium (Ref. 1). There are various methods in reducing the production of minor actinides and plutonium, one of which is the use of fuels with thorium basis in the heavy water reactors. As compared to uranium fuel, thorium fuel has better chemical stability, more appropriate thermal conductivity and higher resistance to proliferation, creating high rate of minor actinides and higher quality of waste. This fuel is also found in the surface of the earth with the rate 3 to 4 times more than uranium fuel. Safety and proliferation resistance, as the advantageous features of thorium fuels, are the factors considered in nuclear technology. Hence, extensive research has been done on it in the countries having extensive amounts of thorium fuel sources, such as Great Britain, Germany, U.S.A., etc. Refs. 2, 3 and 4, 5.

II. Material and methods

Four types of thorium-uranium fuels are applied in the simulations in this study, which are used in order to calculate neutronic parameters, mainly burn up, inventory of MA (minor actinides), variations of reactor poison concentrates, radial and axial fluxes of neutrons, and production of fissile isotope. Calculations were done by Monte Carlo N-Particle eXnded (MCNPX2.6), while CINDER90 code was also used in calculating the burn up. The calculations involved in neutronic parameters apply KCODE that consist 7000 initial neutrons, 50 ineffective cycles and 250 effective ones. Moreover, some experiments involving heavy water zero power reactor are simulated by the Monte Carlo method (MCNPX).

II.A. Introduction to MCNPX and CINDER90 Codes

Los Alamos National Lab. (LANL) established Monte Carlo N-Particle eXended code with 3-dimensional geometry, which is used for calculation of continuous energy transportation based on time for different atomic particles, i.e. neutrons, photons, electrons, or their couples (Ref. 5). This code indicates that the specific values of critical systems can be calculated. The energy region for neutrons is in the range of 10-11 to 20 MeV. Moreover, the reactions in a specific evaluation in a

cross-section involving thermal neutrons are considered in S (α, β) and free gas models. Various applications including nuclear criticality safety, shielding for radiation, burn up, safeguards for nuclear aspects, detector design, design of accelerator, radiotherapy and medical physics use Monte Carlo N-Particle eXended code.

Calculating burn up is concerned with Monte Carlo N-Particle eXended (MCNPX2.6) steady state reaction calculation linked to a deterministic depletion calculation (CINDER90). CINDER90 deals with calculating the depletion for finding out the nuclide densities of the radioactive decay chain per unit time. The nuclide densities, atomic particles emission including neutron and gamma as well as decay constant numbers are applied in finding out the distributions for the energy of particles emission and the delay time. Moreover, CINDER90 is involved with inherent decay and 3400 isotopes within 63 data groups. By taking the new number densities, MCNPX creates new fluxes and reactions, and the procedure is repeated until reaching the end stage Refs.5, 6.

II.B. Heavy water research reactor description and simulation

The heavy water research reactor is a tank-type reactor with pressure tubes, using this reactor includes a tank with pressure tubes that uses moderator, and heavy water coolant, as well as a hexagonal shape lattice that contains fuel bundles with 19 pins, and natural uranium fuel. The devices for empirical purposes in the reactor are a vertical thimble for the central test loop, a medical beam tube with the diameter of 15 cm, and another three beam tubes with 30 mm diameter. Hence, various activities can be done with the mentioned tubes and devices. The activities in this regard include testing the fuel, producing radioisotope, profiling neutron depth, neutron radiography, analysis of neutron activation, etc. It is assumed that various tubes and elements for fuel bundles are of Zircalloy-2, and the center tube filled with coolant has similar diameters (both inside and outside) as the fuel-cladding tubes. The schematic model of reactor, as simulated by MCNPX2.6, is depicted in Fig1.

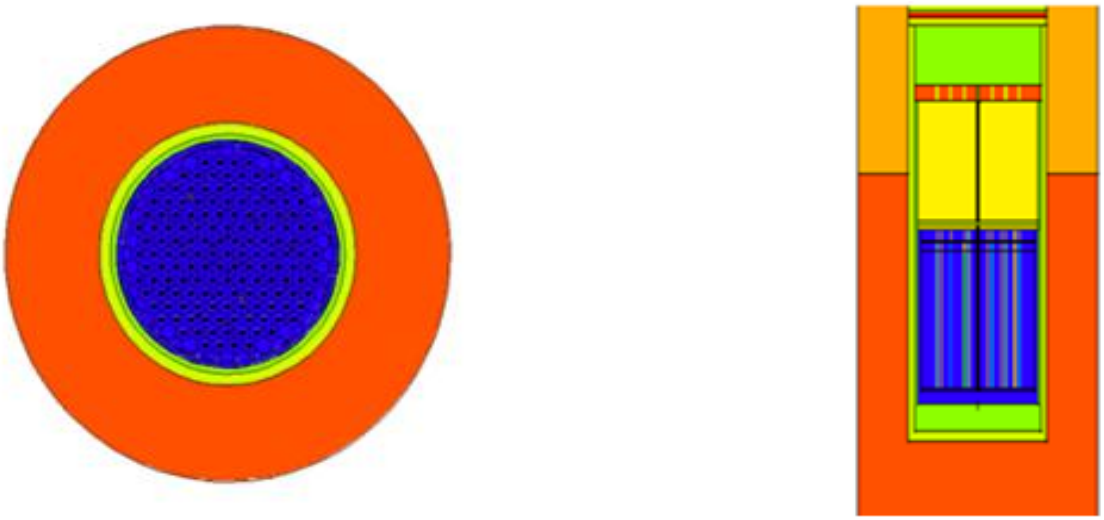

Fig.1: Reactor core layout simulated by MCNPX (Z=0, Y=0)

II.C. Fuel compositions / the right keff

After code benchmark and reactor simulation, the next step was to find the correct fuel compositions. The correct ^{235}U and UO2 weight percent in the fuels were found by varying the weight percent of UO2 in the THO2-UO2 mixture from 4%

to 35%, with different degrees of ^{235}U percent from 5% to 20%. In the first step for each case, fuel theoretical density and neutronic parameter such as keff (multiplication factor), was calculated and are compared with the related data from Uranium fuel. So based on obtained results it was decided to focus on four different fissile fuels components that have nearest neutronic parameters with reference fuel (natural uranium). These fuels are presented in Table I. As specified in the table, because of the multiplication factor depends on the fuel density variation, the amount of fissile isotope, reactor poisons and other factors, so k_{eff} for the fuels varies from 1.017 to 1.13. Fuel A and Fuel C have maximum value of uranium 235 at t=0 so k_{eff} is high for this fuel than the other fuel.

TABLE I Fuel Composition

	Fuel	^{235}U [wt.%]	Density(g/cc)	Keff
A	10%UO2-90%THO2	20	10.3171	1.13770
B	15%UO2-85%THO2	10	10.3176	1.02604
C	30%UO2-70%THO2	5	10.3189	1.06480
D	35%UO2-65%THO2	3.7	10.3194	1.01754
E	UO2	Natural	10.4	1.05231

III. Result

Burn up calculation was done to find out the reduction of plutonium and MA inventory by thorium-uranium fuel in a heavy water research reactor. The results indicated changing in nuclide mas and fuel composition. Neutronic parameters including flux, k_{eff}, etc. as well as the mass balance of the isotope of plutonium and MA inventory were analyzed and compared in thorium-uranium and UO2 fuels.

III.A. Burn-up calculations

III.A.1. Evolution of Multiplication Factor and neutron flux

The evolution of effective multiplication factor (keff) for four thorium-uranium fuels and reference UO2 fuel is shown in Fig. 3. The results indicated that keff for all the fuels decreases with time due to the depletion of the fissile isotopes and the production of the fission products and poisons. The sharp decrease at the beginning is due to the release of 135Xe. 135Xe has a big neutron capture cross section and it has impact on the thermal utilization factor and thus multiplication factor. It is an important poison in reactor operation. 10%UO2-90%THO2 (enrich=20%) fuel, the multiplication factor has a lowest (due to the low consumption of uranium-235) and natural UO2 fuel has highest (due to the high consumption of uranium-235) descending slope during total burn-up time toward to others fuels.

Neuron flux variations during burn up time for all types of fuels have been presented in Fig. 4. The results indicated that neutron flux increase with burn up time that is a consequence of fissile isotope consumes and increase the number of fissions, also the neutron flux value and variations for all of fuels is almost in a same range (10^{13} n/s×cm2) so is completely suitable for research purposes. (15%UO2-85%THO2) and (35% UO2-65% THO2) fuels have a maximum and minimum neutron flux respectively.

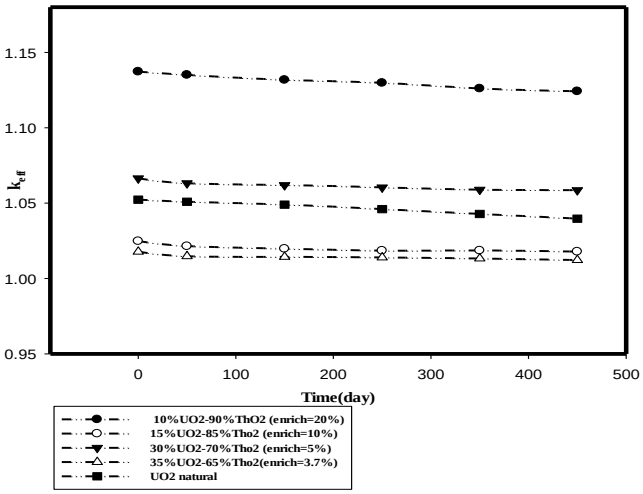

Fig. 3 Multiplication factor (k_{eff}) as a function of burn-up time.

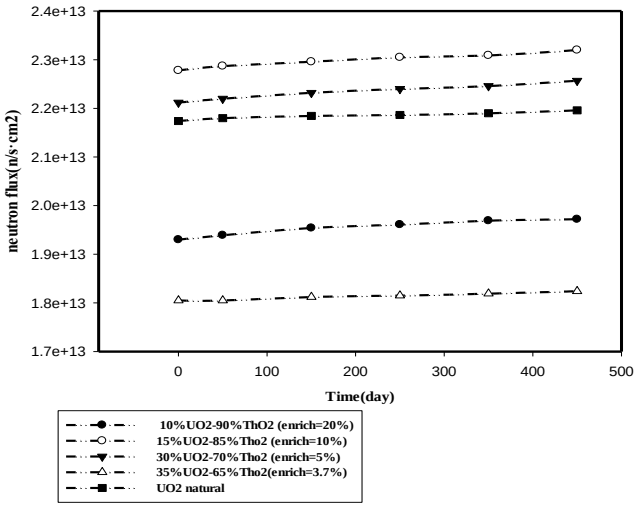

Fig.4 Neutron flux as a function of burn-up time for the different fuel loads

III.A.2. Mass changes of fissile isotopes

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

Fig.5 shows mass changes of 235U isotope as a function of burn-up time. It is clear that after 360 days, the amount of 235U for 10%UO2-90%THO2 fuel has the maximum value (163.2 kg) and the minimum consume (10.4%), while the amount of 235U isotope for natural UO2 has the minimum value (70.2kg) and the maximum consume (21%). Hence, the utilization efficiency for 235U in UO2 fuel is higher than others. Hence, the utilization efficiency for 235U in UO2 fuel is higher than others.

233U has a very small cross section for neutron capture, produced from thorium and often occurs in most fission. This isotope 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 233U 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. Fuel B has lower weight percent of THO2 and higher consume of 235U (higher number of fission) than Fuel A, so the amount of 233U for 15% UO2-85% THO2 fuel is high.

As we know 239Pu is formed through the capture of a neutron in238U and two consecutive β decays of 239U. The mass changes of 239Pu as a function of burn-up and burn-up time are presented in Fig. 7and Fig. 8 respectively. It is obvious in presented figures that the amount of 239Pu mass for natural uranium fuel is very higher than Thorium-Uranium based fuels, Because of the higher percentage of 238U (99.2%) than the other fuels that can produce large amounts of 239Pu.The amount of produced239Pu mass for UO2fuel(11970 gr) in 450 days is about 15.9 times more than 10% UO2-90% THO2 fuel(748.7gr), 8.4 times more than 15% UO2-85% THO2 fuel(1422gr), almost 4.7 times more than30% UO2-70% THO2 fuel(2531gr) and about 3.64 times more than 35% UO2-65% THO2 fuel(3288gr). Also it is clear from Fig. 7 that fuel burn-up for Thorium-Uranium mixed fuels is higher than UO2 fuel.

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 is decreased with increasing of the uranium percentage in the fuels, therefore, it is obvious that 10% UO2-90% THO2 fuel has the maximum burn-up, and natural UO2 fuel has the minimum burn-up.

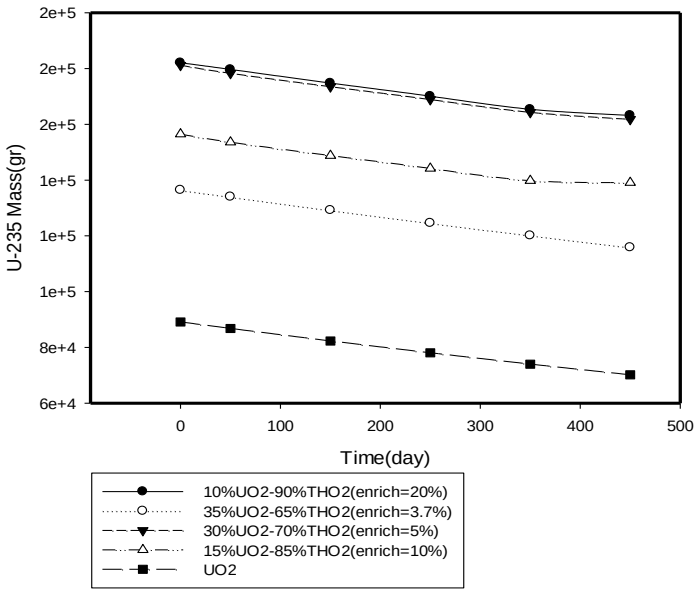

Fig.5: Variation in the mass of U-235 isotope as a function of Burn-up time.

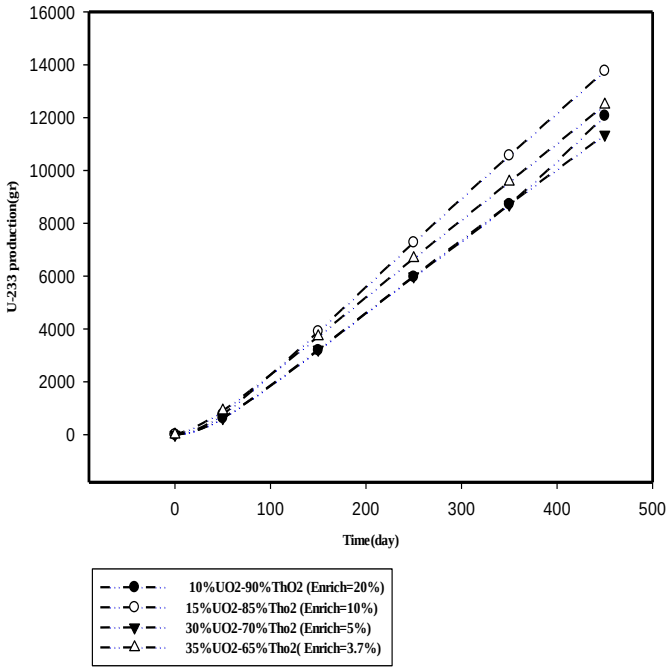

Fig.6: Variation in the mass of U-233 isotope as a function of Burn-up time

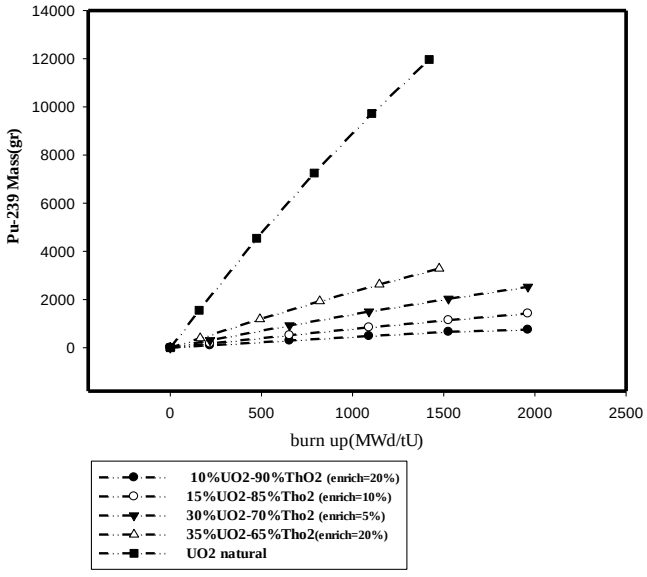

Fig.7: Variation in the mass of PU-239 isotope as a function of Burn-up.

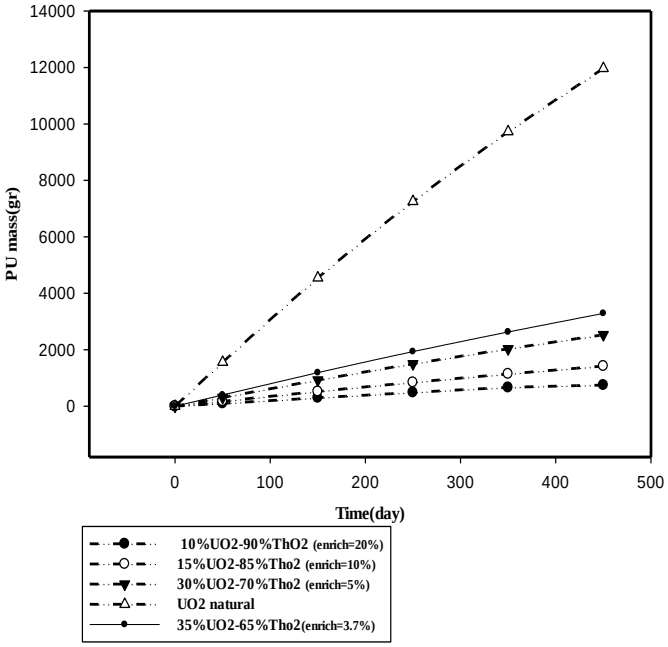

Fig.8: Variation in the mass of PU-239 isotope as a function of Burn-up time.

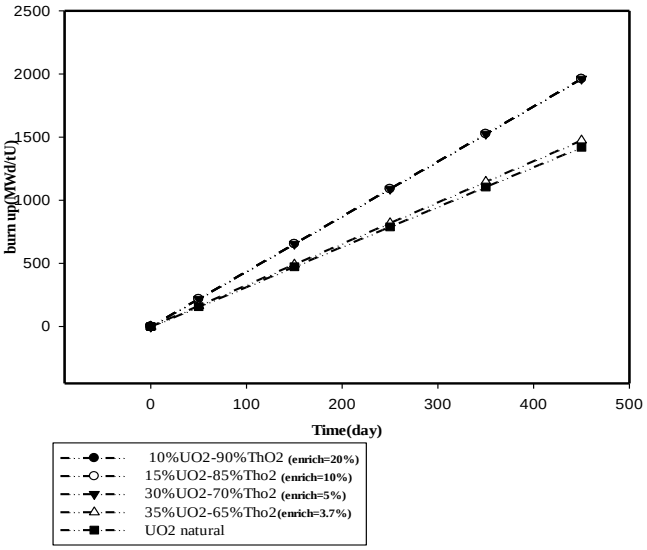

Fig.9: Burn-up behavior as a function of time.

III.A.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 minor actinides, Np, Am, Cm, etc) that have long half-lives and strong α – decaying and is produced based on following reactions:

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

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

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 Fig.10 to Fig.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.

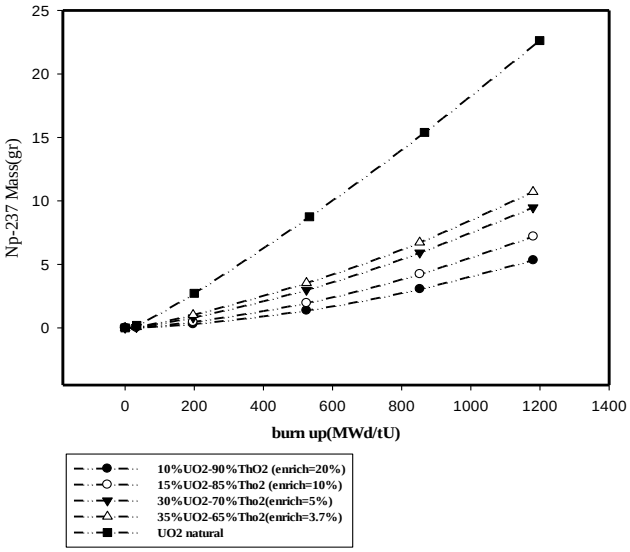

Fig.10: ^{237}Np mass variations for the all of fuels.

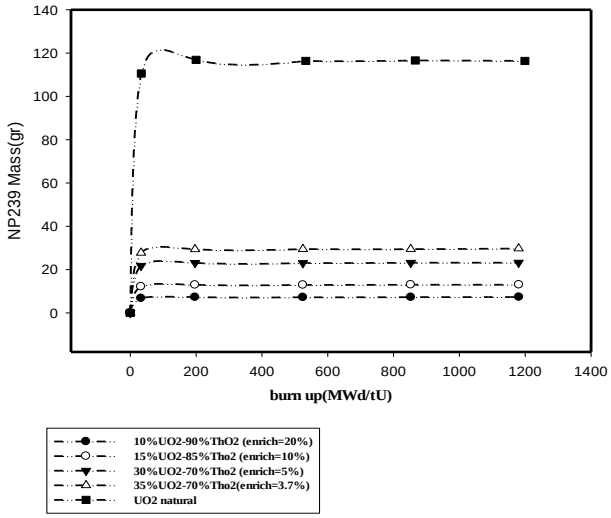

Fig.11: ^{239}Np mass variations for the all of fuels.

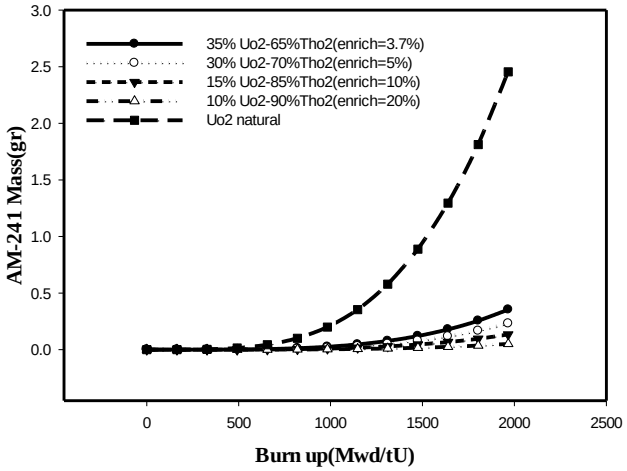

Fig. 12: ^{241}Am mass variations for the all of fuels.

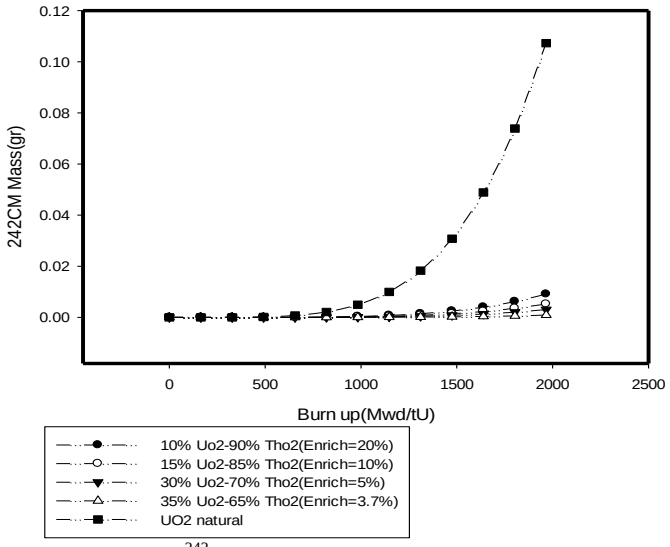

Fig.13: ^{242}Cm mass variations for the all of fuels.

IV. CONCLUSION

Based on investigations regarding the fuel burn up calculations, it can be seen that thorium-uranium fuel is more beneficial to be used in heavy water research reactors, having various advantages as compared to natural uranium fuel, although it also has some disadvantages in comparison. Thorium-uranium fuel can reduce the production of plutonium, long-life of α-emitter isotope (MA), and production of radiotoxic wastes. It also causes reduction of multiplication factor and reactive time. By increasing thorium-uranium fuel burn up, the percentage of uranium in the fuel would be decreased. The fuel (10% UO2-90% THO2) shows a maximum burn up, while natural uranium fuel has a minimum burn up activity, which is an advantage for the thorium-uranium fuel. According to the obtained results, thorium-uranium fuel has a higher weight and lower consumption of 235U. Thus, the efficient utilization of 235U is higher for the uranium fuel.

REFERENCES

[1] J. R, Lamarsh; A. J Baratta. Introduction to Nuclear Engineering, 3rd ed.; Addison-Wesley: New York, USA,2001.
[2] L, Chanyun. MSc, Reactor Physics Department, Royal Institute of Technology, Stockholm, Sweden, 2008.
[3] K. D, Weaver; J. S. Herring. In International Congress on Advanced Nuclear PowerPlants (ICAPP). ANS Annual Meeting; Hollywood, Florida, USA, 2002, PP. 9-13.
[4] I.V, Shamanin.; V.M, Grachev; V.V, Knyshev; S.V, Bedenko; N.G, Novikova. J. Phys.Conference Series, Volume 781, Issue 1,2017.
[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. MCNPX2. 7, Nuclear Engineering and Non-proliferation Division, 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, 2008.