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ThO_2 spent fuel assembly's gamma dose rate dependency to burnup and cooling time

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HIGHLIGHTS

- Thorium-based fuel gamma dose rate dependencies on burn-up values were calculated.
- The required shield thickness was discussed for transportation of the Thorium-based spent fuel.
- Gamma dose rate of the thorium-based and uranium-based fuels were compared in cooling times.

ABSTRACT

Today thorium based fuels are being investigated as possible alternatives in fuel technology. However, the majority of thorium fuel research studies are limited to reactor physics investigations, and there are concerns about dose evaluation and shielding of such spent fuels. The present work investigates burn-up behavior of thorium oxide fuel in Tehran research reactor. The fuel gamma dose rates are calculated at different burnups and cooling times. A comparison between the reactor routine fuel and the thorium oxide fuel is conducted to reveal shielding challenges of the thorium-based fuel application. Despite the U_3O_8-Al routine fuel, the obtained results showed that ThO_2 gamma dose rates are completely dependent on the burn-up values. In addition, for transporting the spent ThO_2 fuel with the routine transport casks, there is a necessity for the higher cooling times compared with that of U_3O_8-Al. Otherwise, construction of thicker transport casks is needed to transport the thorium-based spent fuel in shorter times.

KEYWORDS
Gamma dose rate
Thorium spent fuel
Computational calculations
MCNPX code

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1 Introduction

Thorium-based fuel cycles offer many potential benefits, including greater fuel cycle length due to being breeder in thermal, epithermal and fast neutron spectra, higher explored resources of thorium in the earth crust that are several times higher than the reserves of uranium, as well as improved waste management compared with uranium-based fuels. The use of thorium-based fuel cycles has been the subject of researches for more than 30 years and they were used successfully in the past. In 1960s, the U.S. spent between 5 to 11 billion dollars to produce and separate ~1.55 metric tons of U-233 from irradiated thorium fuel at government weapons material and commercial power reactors. In addition, a molten salt reactor was developed at the Oak Ridge National Laboratory in the late 1960s to demonstrate the potential for breeding U-233. The first commercial nuclear plant to utilize thorium was Indian Point Unit I, a pressurized water reactor that began operation in 1962. The Peach Bottom I Unit, a prototype 40 MW high-temperature gas-cooled reactor used thorium fuel. It operated from 1967 to 1974. Thorium-based fuels can be used in all proven reactor types including PWRs, VVERs, BWRs, HWRs, FBR, HTGR, and in possible future reactor concepts such as molten salt or aqueous homogeneous suspension reactors. Nevertheless, U-232 isotope has attracted a lot of attention in thorium-based cycle. It is generated by means of (n, 2n) reaction taking place on Th-232, Pa-233 and U-233 isotopes. Half-life of U-232 is 69 years. Among its daughter products, there is Tl-208 isotope with very short lifetime emitting hard gamma particles (2.6 MeV). Dose rates in thorium fuel will rise due to accumulation of U-232, which creates additional problems in dealing with spent nuclear fuel of thorium reactors. It should also be mentioned that the surface dose rate from a 55-gallon drum of thorium oxide is approximately 60 mR/h, which is about 13 times higher than a similar-sized drum of uranium (Naymushin et al., 2016; IAEA-TECDOC, 2005, 2000; Wojtaszek et al., 2018). Therefore, higher dose rates of thorium-based fuels

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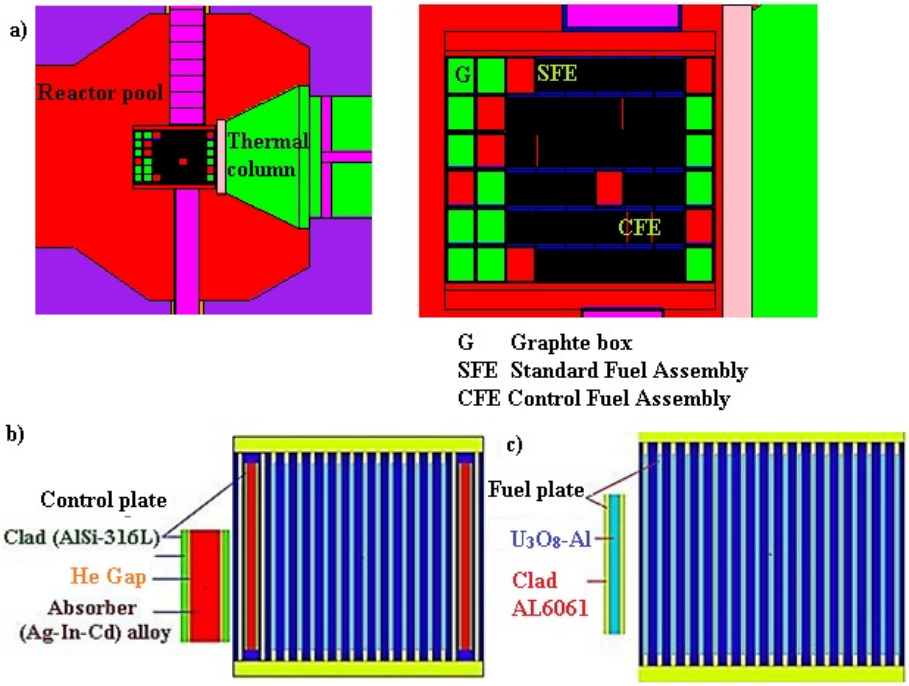

Figure 1: Cross sectional view of a) the modeled TRR core, b) SFE, and c) CFE.

in both fresh and spent situations is a crucial concern and more attention needs to be paid.

Wojtaszek et al. (Wojtaszek et al., 2018) investigated the application of thorium-based fuels as well as their heat power. Their report show that the deployment of the medium-burnup slightly enriched uranium mixed with thorium in Canada would reduce resource consumption by 23% compared with the low burnup NU fuel. They indicated the decay heat power of the fuels from 5 to 40 years after discharge are directly proportional to their burnups (Wojtaszek et al., 2018). Srinivasan et al. (Srinivasan et al., 2006) investigated the gamma dose rate build up with time for a fresh 54 pin advanced heavy water reactor (AHWR) composite cluster containing 500 ppm of U-232 in 1.482 kg of U-233. They used ORIGEN and MCNPX codes to calculate the dose rates. Their results showed the shield requirements increase with time after fabrication, due to the rapid buildup of the hard gamma emitting daughter products of Th-228 in the bundle (Srinivasan et al., 2006). Kang and von Hippel (Kang and von Hippel, 2001) calculated the U-232 gamma-emitter buildup effects on the dose rate at a distance of 0.5 meters (a typical working distance for glove-box operations) from 5 kg spheres of freshly separated U-233 containing 0, 1 and 5 ppm of U-232. Their results showed gamma radiation from U-232 makes the U-233 from high burnup ^{233}U-Th fuel cycles more hazardous than the plutonium in view of radiation protection. The necessity for remote handling of heavily U-232 contaminated U-233 in a closed fuel cycle provides a strong incentive for integration of reprocessing and fuel-fabrication (Kang and von Hippel, 2001). Frybort (Frybort, 2014) reported a comparison of the radiological hazard of thorium and uranium spent fuels from VVER-1000 reactor. Their results show that the dose rate from the spent thorium fuel is increasing in the analyzed period compared with the spent uranium fuel, and it will be greater by almost 15% after 50 years of decay (Frybort, 2014).

The present study investigates thorium oxide dose rates after different burnups in Tehran Research Reactor (TRR).

2 Materials and Methods

The TRR is an open pool, MTR type light water-moderated and cooled reactor with a thermal power of 5 MW. The TRR is operated under critical mode. The reactor core is composed of two types of fuel assemblies: standard fuel elements (SFE) and control fuel elements (CFE). The first considered core (No. 83) consists of 28 SFEs containing 19 fuel plates and 5 CFEs containing 14 fuel plates. The core is fueled with 20% enriched U_3O_8-Al MTR fuels and cooled with the mass flow rate of 500 m^3/h. Two types of control rods are used in the TRR: one made of AgInCd alloy, and the other of stainless steel. Both of them have a set of two control plates as a fork type shape (Mirvakili et al., 2012). A schematic view of TRR core is shown in Fig. 1.

MCNPX 2.6.0 code is used to calculate the fuel assembly dose rates in the air and lead cask (Pelowitz et al., 2005; Fensin, 2008). ENDF.VI library was used for the MCNPX calculations. A lead cask was modeled (Fig. 2) and a number of Geiger Müller dosimeters were placed around the modeled cask at different positions to calculate the gamma dose rate consequence of the spent fuel assembly. ORIGEN code was used to calculate the tho-

rium oxide fuel assembly gamma emission rates on different burnup values and cooling times (Croff, 1980). Libraries of No. 9 for delayed gamma and No. 204, 205 and 206 for calculation of light nuclides, fission products and minor actinides were used by the ORIGEN code input. The gamma source extracted from the ORIGEN code was used in MCNPX code input as a gamma source for the dose rate calculations.

TRR core involves 33 fuel assemblies which maximum received power of the fuel assemblies is 0.15 MW at 5 MW operational power of the research reactor. The value is used to calculate different burnups of the spent fuels using ORIGEN code.

First, the spent thorium oxide fuel dose rates were calculated in the air at different burnups. The dose rates in air were calculated at 100 cm distance from the fuel center. U_3O_8-Al spent fuel dose rate was compared with that of ThO_2 at the same burnups and cooling time. A 20 cm thickness lead cask wall was used and the dose rates of the cask surface when is loaded with the spent thorium oxide fuel of 54 MWD burnup and 150 days cooling was calculated and compared with U_3O_8-Al loading. Heat power of the spent U_3O_8-Al and ThO_2 fuel assemblies of 54 MWD burn-up and 150 days cooling was calculated inside the transport cask and compared with each other. The distribution of the deposited heat and gamma dose rates inside the lead cask was calculated using a mesh tally card of MCNPX code and was calculated for the two different fuels at the same conditions. U-233 buildup in the spent ThO_2 fuel assemblies with different burnups was extracted from the ORIGEN code for the different cooling times and the results was discussed.

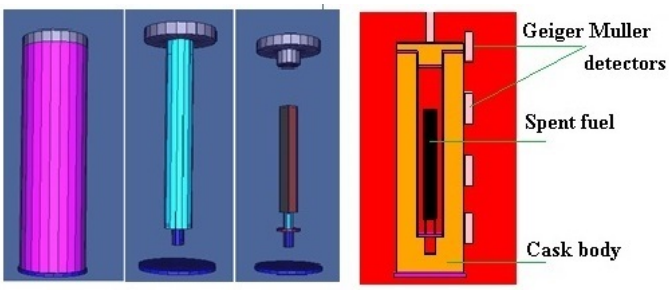

Figure 2: Cross sectional view of a lead transport cask and Geiger Müller dosimeters around the cask. The wall thickness is 15 cm.

3 Results and discussion

3.1 Calculation of the spent ThO_2 fuel dose rates at air

First, the ThO_2 spent fuel dose rates were calculated and compared with each other at different burnup values (4.5 to 54 MWD). TRR full power is 5 MW and the values therefore belong to the loadings of 1 to 12 months, respectively. Figure 3 shows that the dose rates of the spent fuel change from 10^7 mSv/h to 10^9 mSv/h at end of cycle (EOC) which is correspondence with 4.5 to 54 MWD burnups, respectively. The figure shows the dose rate variations at different cooling times up to 100 days. As the figure shows, the dose rates curves of 4.5, 18 and 54 MWD approximately behave in the same way during the cooling times. Another point in the figure is that the dose rates of the ThO_2 spent fuel increases with burnup value growth.

Figures 4 and 5 compare the spent ThO_2 and U_3O_8-Al dose rates at air for 4.5 and 54 MWD burnups at EOC up to 100 days after the fuel cooling time. The calculations show that the low burnup values experience 1.5 to 2 times difference which the gamma dose rates are higher in the case of the spent ThO_2 fuel, while at 54 MWD burnup the differences reach to about 10^2 and 10^3 from the EOC up to the 100 days cooling times. As the figure shows the slope of dose rate falling down of U_3O_8-Al during the cooling times is more than that of ThO_2.

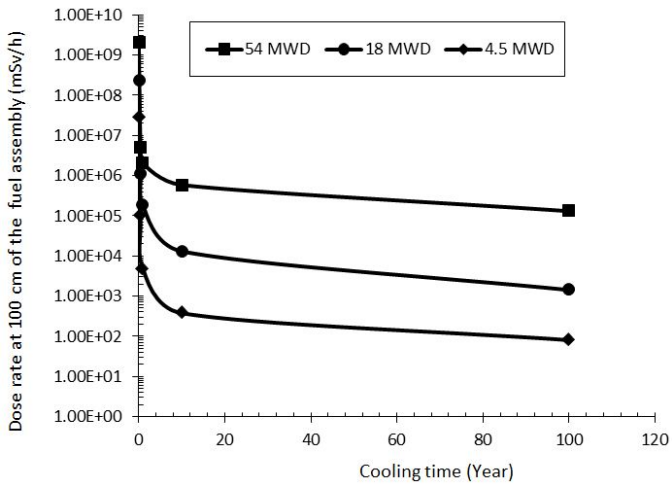

Figure 3: Comparison of the spent ThO_2 fuel dose rate variations at different cooling times and different burnups of the spent fuel. The values belong to 100 cm distance from the fuel surface in air.

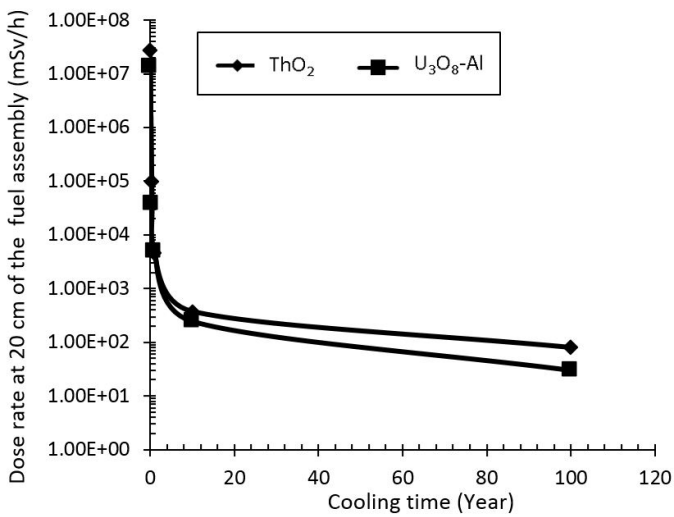

Figure 4: Comparison of the spent ThO_2 and U_3O_8-Al fuel dose rates at air during the cooling times. Fuels burnups: 4.5 MWD.

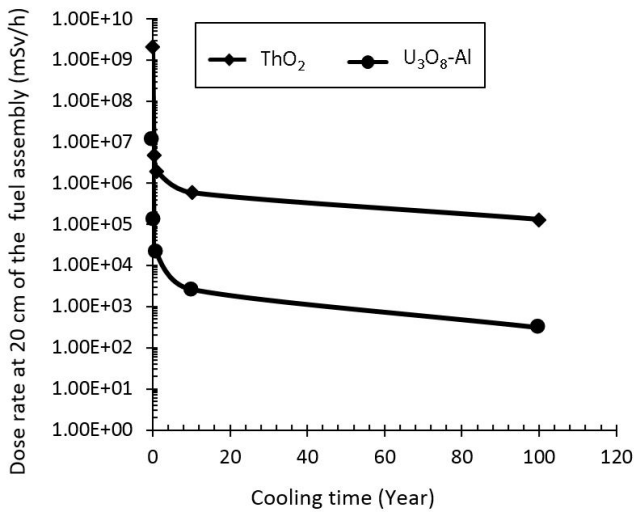

Figure 5: Comparison of the spent ThO_2 and U_3O_8-Al fuel dose rates at air during the cooling times, Fuels burnups: 54 MWD.

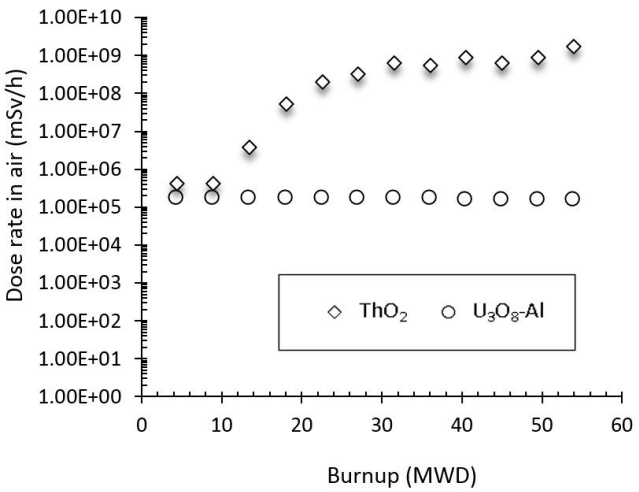

Figure 6: Dependency of the spent ThO_2 and U_3O_8-Al fuel dose rates on burnup values.

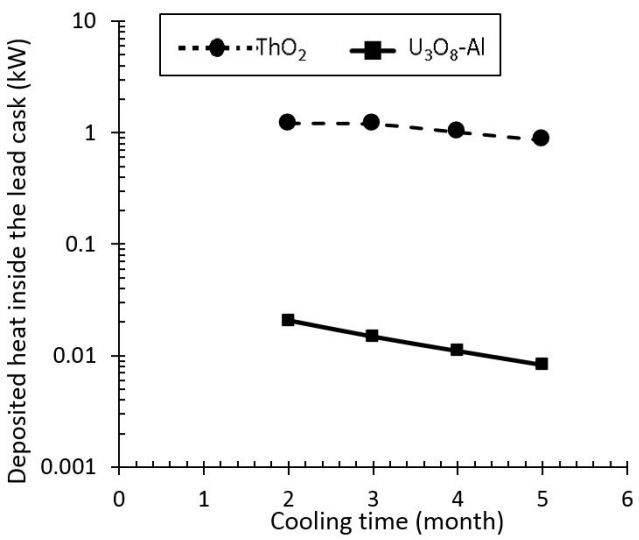

Figure 7: Dependency of the spent ThO_2 and U_3O_8-Al fuel heat power on cooling time.

Figure 6 compares the gamma dose rates of ThO_2 and U_3O_8-Al spent fuel at 100 cm interval of the fuel surfaces at air. As the figure shows, the U_3O_8-Al dose rates are not related to the burnup values, which indicate that the concentrations of the most effective gamma emitter radioisotopes reach to their equilibrium condition after a short burnup. While in the case of the spent ThO_2 the most effective radioisotopes in the gamma dose rates are the long half-life ones which would reach to their equilibrium in long time or high burnups. So it can be claimed that the gamma dose rate emissions of the ThO_2 spent fuel is completely dependent on the fuel burnup values.

Figure 7 shows heat power inside the lead cask after 150 days cooling of the 54 MWD ThO_2 and U_3O_8-Al spent fuels. As the figure shows, the heat power from U_3O_8-Al is considerably less than ThO_2, and during 2 month up to 5 month its value reaches to 8 W, which is one third of its value in the second month of cooling. In the case of ThO_2, the heat power reduction slope is less than that of spent U_3O_8-Al during the same cooling times. In the second month of the cooling time, the heat power from ThO_2 spent fuel loaded inside the lead cask is about 1.21 kW, which the value reaches to about 850 W after passing three months (the 5th month).

Routine casks of the spent U_3O_8-Al have 15 cm thickness and the spent fuels could be carried with them after 3 months of the cooling time, the time that the cask surface dose rates reach to less than 2 mSv/h (USNRC, 1997). Figure 8 compares the deposited heat distribution inside a 20 cm thick lead cask when it contains two different fuels having 54 MWD burn-up and 150-days cooling times. As the figure shows, after 5 months of the cooling time the maximum deposited heat density is 0.066 W/cm^3 in the case of the 54 MWD spent fuel, which would not a major concern for transporting the spent fuel.

Figure 9 compares the two spent fuel's gamma dose rate distribution inside the lead cask in vertical view. As the figure shows, the detector No. 2 which is placed in the hottest section of the cask surface shows 1.6±0.32 mSv/h dose rate in the case of the spent thorium-based fuel loading while the value is 7.8±2.3 μSv/h in the case of the uranium-based fuel loading.

Figure 10 compares the radial gamma dose rate distribution inside the lead cask when it contains the 54 MWD thorium-based spent fuels with two different cooling times. Figure 10-a shows the 90-days cooled fuel which was placed inside the 20 cm lead transport cask. According to the figure, the gamma dose rate at the cask surface is about 44±15 mSv/h. Figure 10-b shows the dose rate distribution around the 15 cm thick lead cask when it involves a 54 MWD and 150-days cooled thorium-based spent fuel. According to the figure, the cask surface dose rate is 16±3.2 mSv/h.

Burnup calculations show that U-233 buildup will occur inside the spent ThO_2 during the different cooling times because of Pa-233 (half-life 27 days) decay to U-333. In the case of 22.5 MWD spent ThO_2 fuel, at EOC the U-233 value is about 35 g and after 1-year cooling time, the value reaches to about 105 g. The issue is important regarding to critically accident concerns in storage casks

(Fig. 11).

All of the calculation errors were less than 2%.

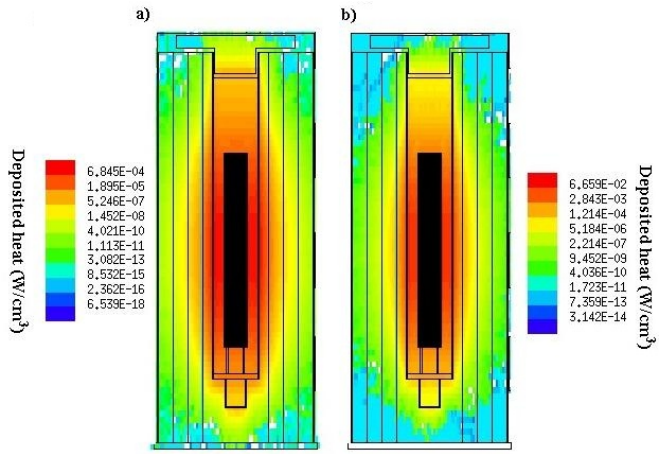

Figure 8: Comparison of the spent ThO_2 and U_3O_8-Al fuel deposited heat distribution in the 20 cm thick lead cask. a) Uranium fuel loading b) Thorium fuel loading.

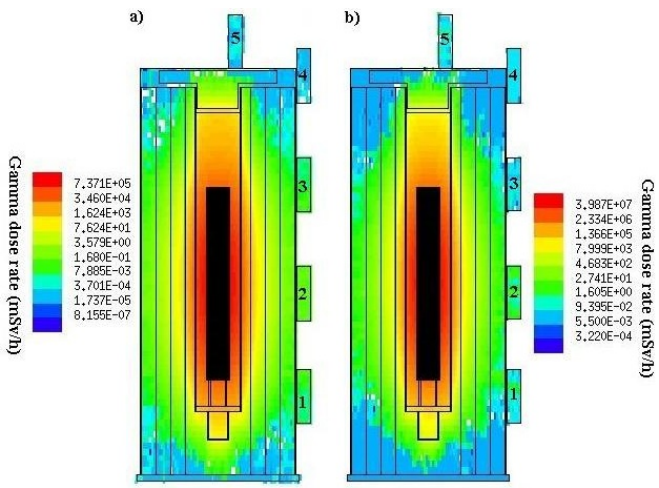

Figure 9: Comparison of the spent ThO_2 and U_3O_8-Al fuel gamma dose rate distribution in the 20 cm thick lead cask a) Uranium fuel loading b) Thorium fuel loading.

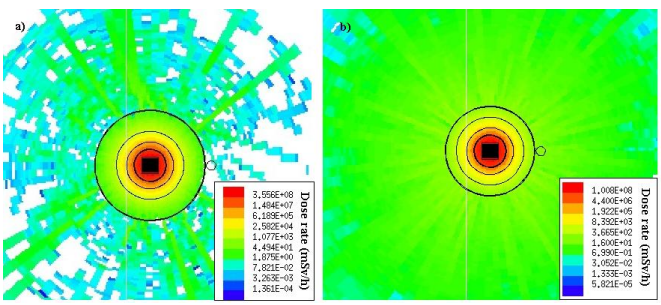

Figure 10: The spent ThO_2 fuel gamma dose rate distribution around the lead cask in cross-sectional view a) after 90 days cooling with 20 cm thick lead cask, b) after 150 days cooling with 15 cm thick lead cask.

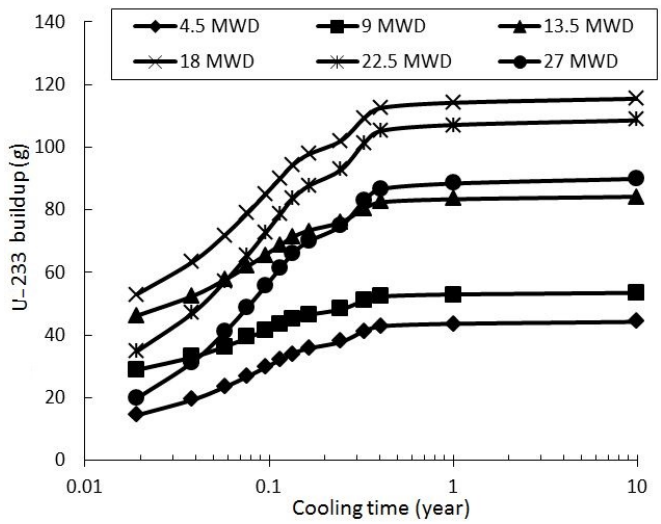

Figure 11: U-233 buildup inside the spent ThO_2 fuel on the cooling time.

4 Conclusion

Thorium fuel has been taken in attention recently because of its satisfied neutronic performances in thermal and fast reactors as many investigations were conducted on these kinds of fuels for application in research and power reactors. However, some literatures pointed to the spent thorium fuel high gamma dose rates, but there is a gap to reveal a precise behavior of the thorium-based spent fuels during their cooling times. The present study shows the thorium-based spent fuels have significantly higher gamma dose rates in comparison with the TRR routine spent fuel (100 to 1000 times, depending on its burnup and cooling time). The results show that only for the burnups less than 15% the differences are less than the previous mentioned values. Therefore, transportation of the thorium-based spent fuel needs thicker lead transport casks if the fuel is to be transported at the same cooling time of the TRR routine spent fuels (3 months). Doubling of the transport time is another solution, which can be used to transport the thorium-based spent fuel with the TRR standard casks.

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