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Investigation of ^{244}Cm dynamics production in a WWER-1200 reactor for fuel based on ^{235}U and MOX fuel

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^{244}Cm is a very important nuclide due to the high specific heat release and it may become reasonable to recycle the spent nuclear fuel to extract this isotope. In this work, the dynamics of ^{244}Cm isotope production for the fuel assembly of the WWER-1200 reactor with an enrichment of 4.95% and burnup up to 70 MW · day/kg was calculated using Serpent Monte Carlo code. A similar calculations was performed for MOX-based fuel assembly with the same irradiation characteristics. It has been shown that production of ^{244}Cm in high burnup uranium dioxide fuel, and especially in MOX fuel, reaches a high value, which can cause problems during spent fuel management. The main problems associated with the curium relates to the residual heat and neutron activity.

Keywords: curium-244, spent fuel, WWER-type ractors, high burnup, residual heat.

Introduction

During operation of a nuclear reactor nuclear fuel is exposed to a high-density neutron flux [1]. ^{238}U contained in large quantities in the uranium dioxide fuel captures neutrons. The resulting products also continue to capture neutrons, which leads to the production of a significant number of nuclides with mass numbers of 239 and above. These nuclides have a high impact on the neutron-physical characteristics of the nuclear fuel, especially when studying the issues of spent nuclear fuel (SNF) management. In particular, ^{239}Pu and ^{241}Pu are the main isotopes producing in the fuel and providing a significant increase of the fuel cycle duration [2].

For many decades the fuel burnup did not exceed 50 MW · day/kg. Only in the recent years, fuel of a new reactors types began to reach burnup of about 70 MW · day/kg. A relatively small increase in the fuel burnup (about 40%), as shown in calculations, can lead to a significant increase in the actinide production (several times and more). The calculations presented in this work shows that ^{244}Cm in the fuel with high burnup (especially in the MOX fuel) will have one of the defining values for the residual heat amount in the SNF (this leads to a serious problem associated with the SNF management). ^{244}Cm is a very important nuclide, due to the high specific heat release [3], and it may probably become reasonable to recycle the SNF to extract this isotope for its further usage.

Curium-244 isotope production scheme

^{244}Cm isotope in thermal-neutron reactors is almost completely accumulated from ^{238}U by capturing six neutrons sequentially. A simplified scheme (it does not show β-decays and fission reactions) of the ^{244}Cm during operating time is presented in Figure 1.

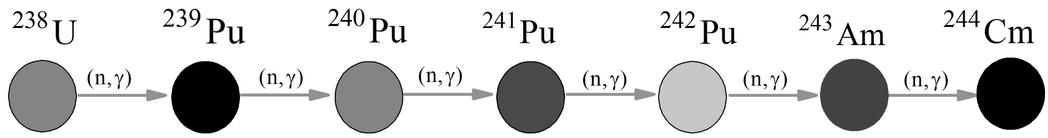

Figure 1. ^{244}Cm isotope production.

The production rate of each successive nuclide depends on the actual amount of the preceding nuclide. Thus, a significant rate of the particular nuclide production reaches only after building up the entire chain of its predecessors. This leads to the fact that the nuclides such as ^{244}Cm (with chains similar to shown in Figure 1) have a strongly nonlinear production dynamics.

The amount of ^{244}Cm produced in a thermal nuclear reactor with uranium dioxide fuel is almost completely determined by the burnup value. The burnup dynamics have a little impact on the result due to the fact that all nuclides in the chain have a half-life significantly longer than the duration of the fuel cycle. ^{238}U isotope concentration in uranium dioxide fuel is about the same for different types of fuel assemblies and is only slightly dependent on burnup.

Calculation model

Serpent Monte Carlo code was used for burnup calculations [4]. The computational model consists of a single fuel assembly of the WWER-1200 reactor with reflective boundary conditions and ^{235}U enrichment of 4.95% [5, 6]. ENDF/B-VII [7] library of evaluated nuclear data was used in this calculations. Model of fuel assembly is shown in Figure 2.

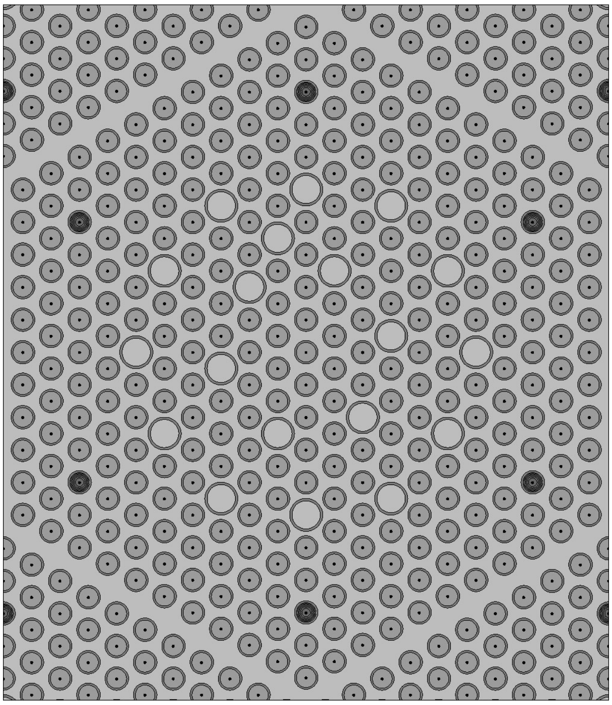

Figure 2. Model of WWER-1200 fuel assembly with reflective boundary conditions in Serpent code.

Calculations were carried out for burnups from 0 to 70 MW $\cdot$ day/kg. This burnup values is a preliminary estimate for the WWER-1200 fuel [8]. For standard types of WWER-1000 fuel assemblies, the maximum burnup value was about 40 MW $\cdot$ day/kg [9] (61.8 MW $\cdot$ day/kg using experimental fuel assemblies and up to 72 MW $\cdot$ day/kg for single fuel elements [10, 11]).

Cm-244 production in thermal nuclear reactors with uranium dioxide fuel

Obtained dependence of the specific content of ^{244}Cm depending on the burnup is shown in Figure 3.

As can be seen, at the initial stage of the fuel cycle and up to a burnup of 20 MW $\cdot$ day/kg, the ^{244}Cm isotope is practically not produced. This is due to the formation of the intermediate nuclides in the chain, after which curium production begins to occur relatively intensively.

In the fuel with burnup of about 40 MW $\cdot$ day/kg the amount of ^{244}Cm produced during operation is several tens of grams per ton and does not make a significant contribution to the radiation or thermal characteristics of the SNF. In this sense, the WWER-1200 reactor with burnup up to 70 MW $\cdot$ day/kg becomes the first practically used reactor type in which the problem of curium is relevant.

It is important to note that the only channel for reducing the amount of ^{244}Cm in a nuclear reactor is the reaction of radiation capture (except for α-decay, with

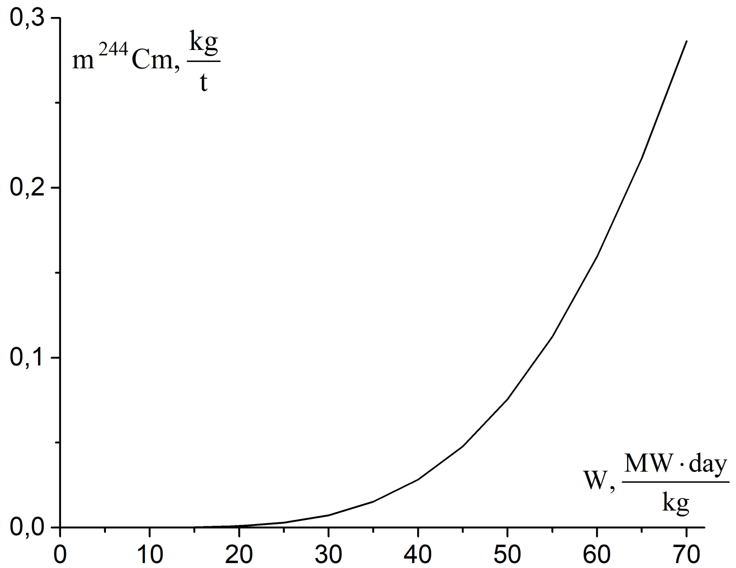

Figure 3. Production of ^{244}Cm depending on burnup for the WWER-1200 reactor fuel assembly.

a half-life of 18 years) with a very low cross section of about 10 barn for thermal neutrons [3]. For this reason, the amount of ^{244}Cm in reactor core decreases very slowly.

Cm-244 production in thermal reactors with MOX fuel

The rapid accumulation of ^{244}Cm in uranium dioxide fuel after building up the chain of precursors (Figure 1), starting from burning out 20 MW · day/kg and above suggests a high rate of production of this isotope in MOX fuel, where this chain is already built. To estimate the ^{244}Cm production time in MOX fuel, a similar model was created for a fuel based on uranium oxide, but using MOX fuel. The composition of MOX fuel was determined as follows: instead of enrichment natural uranium is diluted with plutonium and the $^{239-242}Pu$ isotope proportion that which is contained in the spent uranium fuel of the WWER-1200 reactor at the end of the fuel cycle (the assumption is based on the potential spent fuel of the WWER-1200 reactor management with the emission of plutonium from it). The amount of plutonium in MOX fuel was calculated on the basis that the resulting concentration of fissile isotopes (^{235}U, ^{239}Pu, ^{241}Pu) is the same as in uranium fuel with an enrichment of 4.95%. MOX fuel consist of uranium and plutonium dioxide: UO_2 and PuO_2. The composition of the fuel thus obtained is presented in Table 1.

Table 1.
Mass fraction of % uranium and plutonium isotopes in MOX fuel.

Isotope	^{235}U	^{238}U	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Mass Fraction, %	0.698	92.7	3.29	1.46	1.19	0.678

Production of ^{244}Cm isotope depending in the fuel burnup (composition of fuel in Table 1) is shown in Figure 4.

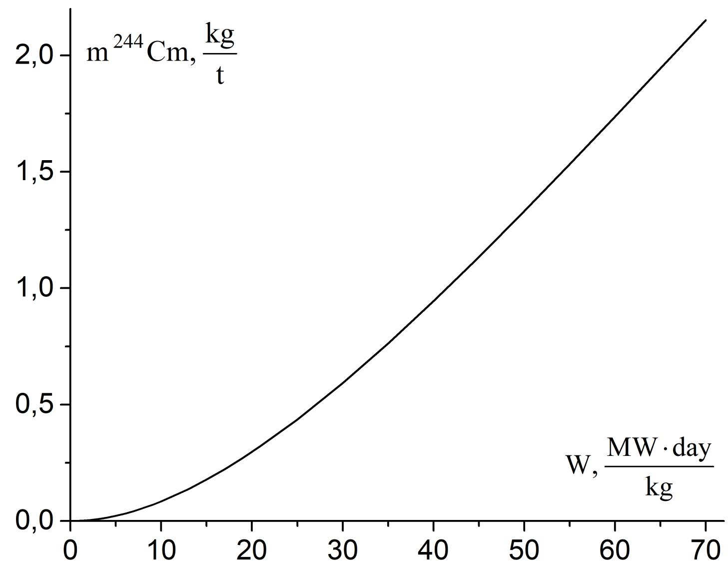

Figure 4. Production of ^{244}Cm depending on burnup in MOX fuel.

As can be seen, ^{244}Cm production in MOX fuel can reach a significant value up to kilograms per ton. Even with relatively small burnup's characteristic of the most WWER-type reactors, the mass of ^{244}Cm reaches 1 kg per ton.

Residual heat and neutron activity

High decay energy in combination with a relatively short half-life leads to the fact that ^{244}Cm creates a high heat release of ~ 2.83 kW per 1 kg of ^{244}Cm. Figure 5 presents the residual heat of 1 ton of spent nuclear fuel from WWER-1000 and WWER-1200 reactors in comparison with the heat generated by 1 kg of ^{244}Cm [12].

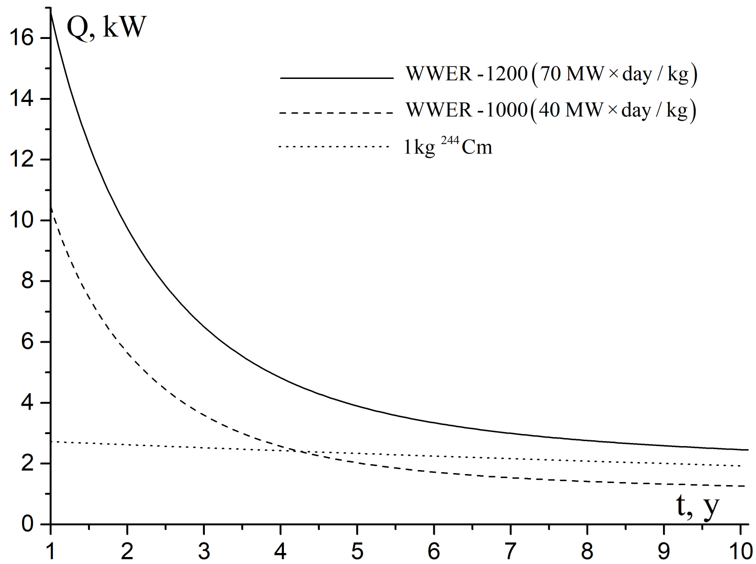

Figure 5. Residual heat of 1 ton SNF from WWER-1000 and WWER-1200 reactors in comparison with heat generated by 1 kg ^{244}Cm.

On the other hand, ^{244}Cm is an important isotope due to the high heat release over a long period of time. ^{228}Pu used in the manufacture of electrical energy sources in space technologies [13-15] have a similar property.

The extraction of curium isotopes for subsequent use is a feasible task with the prospect of further development [16]. For example, in work [13] was discussed the issue of separating curium from spent fuel with a burnup of 33 MW · day/kg in which the production of ^{244}Cm was only 31.9 g/ton. At the same time, in the WWER-1200 reactor with uranium dioxide fuel this value is higher by an order. In the future, the curium production time can reach kilograms per ton by using MOX fuel.

Another important fact is that the ^{244}Cm isotope creates high neutron activity in the SNF due to spontaneous fission. The neutron activity of one gram of ^{244}Cm is $\sim 1.6 \cdot 10^7$neutr./s [17]. ^{244}Cm also makes a significant contribution to the neutron activity due to reactions (α, n) on oxygen ^{17}O and ^{18}O contained in the nuclear fuel [18].

Conclusion

The latest generations of light-water reactors with high burnup fuel (from 60 MW · day/kg and above) pose new challenges for researchers related to the spent nuclear fuel management. One of these tasks can be the solution of the problem with significantly increased ^{244}Cm isotope production. The mass of this isotope in the fuel with a burnup of 70 MW · day/kg exceeds the corresponding mass at a burnup of 40 MW · day/kg by about 6 times.

At the same time the small amount of ^{235}U isotope in natural uranium (is approximately 0.7%) require use of new types of fuel in the future. Usage of MOX fuel can be one of the solution for this problem. But as was shown by calculations it can cause serious problems with spent fuel management.

References

[1] M. Stacey Weston, Nuclear Reactor Physics (Second edition) (Wiley-VCH, 2007) 735 p.
[2] J.J. Duderstadt, L.J. Hamilton, Nuclear Reactor Analysis (WILEY, 1976) 650 p.
[3] JEFF-3.3 Radioactive Decay Data.
https://www.oecd-nea.org/dbdata/jeff/jeff33/index.html#_downloads.
[4] J. Leppaenen, Serpent - a continuous-energy Monte Carlo reactor physics burnup calculation code VTT technical research centre of Finland (2013) 164 p.
[5] Y. Semchenkov, International Nuclear Forum Bulgarian Nuclear Energy "National, Regional and World Energy Security", Varna (2010).
[6] Yu. Dragunov, 5th International Scientific and Technical Conference "Ensuring Safety of NPPs with WWER", Podolsk, (2007)
http://www.gidropress.podolsk.ru/files/proceedings/mntk2007/disc/autorun/article140-en.htm.
[7] ENDF/B-VII.0: Nucl. Data Sheets **107** (2006) 2931.
[8] T. Korbut, Nuclear Physics and Engineering **8**(2) (2017) 118-122.

[9] V. Kolobashkin, Radiation characteristics of irradiated nuclear fuel (M: Energoatomizdat, 1983) 384 p.
[10] O. Samoilov, Proceedings of the 5th International Scientific and Technical Conference "Ensuring the safety of NPP with WWER", Podolsk (2007) http://www.gidropress.podolsk.ru/files/proceedings/mntk2007/disc/autorun/article123-en.htm.
[11] V. Molchanov, 6th International Conference on WWER Fuel Performance, Modeling and Experimental Support., Albena (2005).
[12] A. Piatrouski et al., J. Phys.: Conf. Ser. **1133** (2018) 012009.
[13] C.O. Maidana, Nuclear Technologies for Space Exploration: An overview on nuclear thermal and electric propulsion, radioisotope power generators and fission surface power (Review of the Italian Mars Society, 2014).
[14] A. Postulatov, 5th European Conference on Thermoelectrics, Odessa (2007) http://ect2007.its.org/ect2007.its.org/proc-contents.html.
[15] N. Bibler, The Journal of Physical Chemistry **78**(3) (1974) 211-215.
[16] J. Posey, Process for the recovery of Curium-244 from nuclear waste (ORN L-5687, 1980) 104 p.
[17] EXFOR: International Collaboration Between Nuclear Reaction Data Centres (NRDC), Nucl. Data Sheets **120** (2014) 272.
[18] M. Hiroyuki, Data Book for Calculating Neutron Yields from Reaction (α, n) and Spontaneous Fission, Japan Atomic Energy Research Inst. (1992) 268 p. (In Japanese)