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Original Article

Monte Carlo analysis of LWR spent fuel transmutation in a fusion-fission hybrid reactor system

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ABSTRACT

The aim of this paper is to determine neutronic performances of the light water reactor (LWR) spent fuel mixed with fertile thorium fuel in a FFHR. Time dependent three dimensional calculations for major technical data, such as blanket energy multiplication, tritium breeding ratio, cumulative fissile fuel enrichment and burnup have been performed by using Monte Carlo Neutron-Particle Transport code MCNP5 1.4, coupled with a novel interface code MCNPAS, which is developed by our research group. A self-sustaining tritium breeding ratio (TBR>1.05) has been kept throughout the calculations. The study has shown that the fissile fuel quality will be improved in the course of the transmutation of the LWR spent in the FFHR. The latter has gained the reusable fuel enrichment level conventional LWRs between one and two years. Furthermore, LWR spent fuel - thorium mixture provides higher burn-up values than in light water reactors.

1. Introduction

Fission-based nuclear energy creates environmental problems with nuclear waste in form of higher actinides being the most dangerous ones due to their high and long term activities, followed by fission fragments and radio activated structures. On the other hand, fusion energy is significantly cleaner than fission energy. However it is a long way until pure fusion reactors with high gain factor can become economically competitive. Fusion-fission hybrid reactors (FFHR) consist of a fusion reaction chamber covered with a fissionable blanket. The high energetic 14 MeV (D,T) fusion neutrons produced in the chamber can multiply the fusion power, produce fissile fuel and transmute the nuclear waste of fission reactors simultaneously. FFHR's are seen as the intermediate goal in the development of fusion reactors. The reason for this is that the value of fusion power gain of FFHR's are much lower than pure fusion reactors [1]. From the mid-1950 to up till now, a great number of FFHR designs have been appeared in the literature, worldwide. In the course of the development of fusion nuclear technology and in designing of the FFHR's, neutronic calculations play the important role. Generally, deterministic and stochastic methods are used for the neutronic analysis of FFHR's. The most commonly used deterministic methods are spherical harmonics method (P_L) and discrete ordinates (S_N) methods. ANISN [2] and SCALE [3] nuclear codes solve the Boltzmann transport equation using coupled S_N-P_L method. The Monte Carlo (MC) simulation is the most commonly used stochastic method for solving the neutron transport equation. The interface code named ERDEMLI [4] has processed the output of the ANISN code to evaluate technical data for reactor design and to carry out time evolution analyses for FFHR designs in a great number of works [5–21]. In subsequent studies, the time independent neutron performance of various structural materials has been examined with the MCNP code [22–24]. Recent studies have been focused on time-dependent three-dimensional analysis using MCNP code. Kotschenreuthera et al. have been investigated nuclear waste conversion in FFHRs. They have performed calculations with MCNPX and ORIGEN codes. They concluded that waste disposal in the FFHR's is more economical than other alternative methods [25]. Matsunaka et al. have been calculated burn up value using MCNP and ORIGEN Codes and have developed a new method to regenerate neutron cross sections [26,27]. Ma et al. have been performed neutronic analyses using the MCNP and BISONC codes on the thorium-based FFHR blanket. They have obtained high cumulative fissile fuel enrichment and multiplication factors [28]. Zhou et al. have been studied fuel cycle in fission blanket using the MCNP and ORIGEN codes. They have

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calculated 12 times fuel charge change during the 60 years of plant operation [29]. Zheng et al. have been performed neutronic analysis in PWR spent fuelled FFHR. They have been used MCNP and ORIGEN codes in calculations. They have evaluated high multiplication factor (M) and tritium breeding ratio (TBR) values [30]. Zu et al. have developed a code system to perform neutronic analysis in FFHR with pressure tube type blanket. The code system consists of DRAGON code and multi group MC code [31]. Siddique et al. have analyzed waste recycling performance of the FFHR using MCNPX and MONTEBURNS codes [32,33]. The well-known and widespread time calculations codes, coupled with MCNP make one group point kinetic time calculations, such as ORIGEN and MONTEBURNS. These assumptions are obviously very crude. Therefore great discrepancies have been observed by calculations with MONTEBURNS for the TBR value [33]. Xia et al. have investigated neutronic performances in natural thorium and uranium mixture fuelled FFHR with molten salt coolant. They carried out their calculations with MCNPX and ORIGEN codes and attained high fissile fuel production rate and TBR value [34]. CINDER works with 69-group library, but is still a point kinetic code.

The spent LWR fuels contain higher actinide isotopes in form of reactor grade plutonium (RG-Pu) and minor actinides (MA) as well as highly radioactive and radiotoxic fission products. These topics were subject of various studies, where also the possibility of using mixed fuel made of WG-Pu, RG-Pu and MA in form of mixed fuel with thorium has been investigated intensively [7–34].

Another way to think about the incineration of highly radioactive waste with thorium is the accelerator-driven systems (ADS), which is a conceptual design. Firstly, Carminati et al. have designed a system called energy amplifier. They have been considered that thorium as breeding fuel in this device [35]. After that, Rubbia et al. have proposed an energy amplifier operating with fast neutron. They have asserted that incineration of nuclear waste with thorium provides many practical advantages [36]. In other papers, Rubia et al. have implied that five energy amplifier is the effective solution for the elimination of the LWR waste stockpile in 37 years [37]. These basic concepts have leaded to today's ADS researches. Today, many countries have focused their research on the development of ADS. India's ADS development program mainly concerned about the utilization of thorium using ADS [38]. Another important ADS project is MYRRHA that is coordinated by Belgian Nuclear Research Centre. It has been designed for working both subcritical and critical mode [39]. Barlow et al. have analyzed fuel mixtures that contain thorium in a MYRRHA. The results have shown that MYRRHA is not the solution for long-term waste problem but it has proven conversion ability of the waste isotopes [40,41]. Many conceptual studies have been continuing about the development of ADS.

Institute of Nuclear Energy Safety Technology (INEST/FDS Team), Chinese Academy of Science is the main power which is devoted to the research area of nuclear technology, safety and material which are the common and crucial technology among various fusion and fission [43–45]. In the last 30 years, series of lead-based reactors (FDS and CLEAR series) was developed not only for fusion, but also for advanced fission energy, such as fast reactor systems, small module reactor, and accelerator-driven system (ADS). Significant achievements on technology R&D have been reached. For example, lead alloy coolant loops (DRAGON and KYLIN series) and integrated test facilities (CLEAR-0, CLEAR-S, CLEAR-V) were built with the advanced experimental functions and operating parameters which can validate the key technologies of blankets for ITER-TBM, CFETR, DEMO, as well as lead-based fission reactor; the high intensity neutron generator HINEG has produced the D-T fusion neutrons with the yield up to 6.4×1012 n/sec [46], which is a highlight in the field of nuclear science and technology in recent years, and will significantly promote the development and validation of advanced nuclear systems.

The available time calculations codes are based on point kinetic models and hence inappropriate to follow the conversion and transmutation of isotopes in the fuel zone by zone for such an advanced code like MCNP. Therefore, it is necessary to develop new code(s) for time dependent solutions of Boltzmann Transport Equation at appropriate/comparable level with MCNP. For this reason, a novel interface code has been developed to perform time-dependent neutronic analyses with MCNP code. This interface code reads the MCNP output and calculates the variation of the densities of the selected isotopes by neutron interactions and radioactive decay, creation of new isotopes in discrete time steps and creates a new input file for the MCNP throughout the reactor operation time. In parallel, it calculates neutronic parameters, which are not included in the MCNP package, such as energy multiplication factor (M) for FFHR, cumulative fissile fuel enrichment (CFFE), net fissile fuel regeneration ability of reactor in the form of figure of merit (FOM), isotopic changes in fuel element and fuel burn-up. The present work investigates the potential of the utilization and transmutation of spent nuclear fuel mixed with thorium in hybrid reactors. For that purpose, an auxiliary novel interface code has been developed for MCNP5 v1.4 [42] to achieve time dependent neutronic analysis, described in chapter 2.

The principal objective of the study is to assess the performance of a FFHR in 3-D modeling with code (MCNP) to incinerate LWR spent fuel through additional energy production in conjunction with the implementation of substantial amount of thorium into the energy vector. The development of an advanced time evolution code for that purpose came as an important outcome.

2. Calculation methods and problem description

The interface code has been written in FORTRAN 90 computer programming language and is called MCNPAS (MCNP Assessment Code) [47]. The flow chart of the interface code to MCNPAS process the MCNP output is seen in Fig. 1. The main task of the interface code is to trace isotopic changes formed due to the nuclear reactions and radioactive decay. Fig. 2 depicts the transformation schema of the actinide isotopes, adopted from Ref. [18] and imbedded in MCNPAS. The interface code is run in sequence using the Windows batch file. In the first step, the MCNP code calculates the neutron fluxes and reaction rates in the cells and surfaces. In the second step, the interface code reads required data from output and input file of the MCNP. Thereafter interface code calculates neutronic performance parameters and prepares new input file for the MCNP.

The calculations and isotopic changes have been made in a user defined discrete time steps. The reactor power, plant factor (PF), discrete time steps (Δt), the areas and volumes of the regions in FFHR blanket, neutron load in the first wall are given as input. In this study, all calculations have been made for an energy flux of 2.25 MW/m^2 at the first wall corresponding to a neutron wall load of 10^{14} $n/(cm^2 sec)$ of 14 MeV neutrons under consideration of a plant factor of 100% for a deuterium-tritium fusion reactor. The neutronic parameters have been followed during four years of operation period.

The neutronic calculations have been performed for an experimental fusion fission hybrid reactor blanket concept shown in Fig. 3, and described in Refs. [48,49] in detail. The first wall and the cladding material of rods are made of SS316L stainless steel. The tritium breeding materials are the molten salt FLIBE (Li_2BeF_4) in the fuel zone, which acts also as coolant and Li_2O zones beyond the fuel zone. The Li_2O and graphite reflector zones are made in sandwich structure, as suggested in Ref. [48]. The first and second graphite

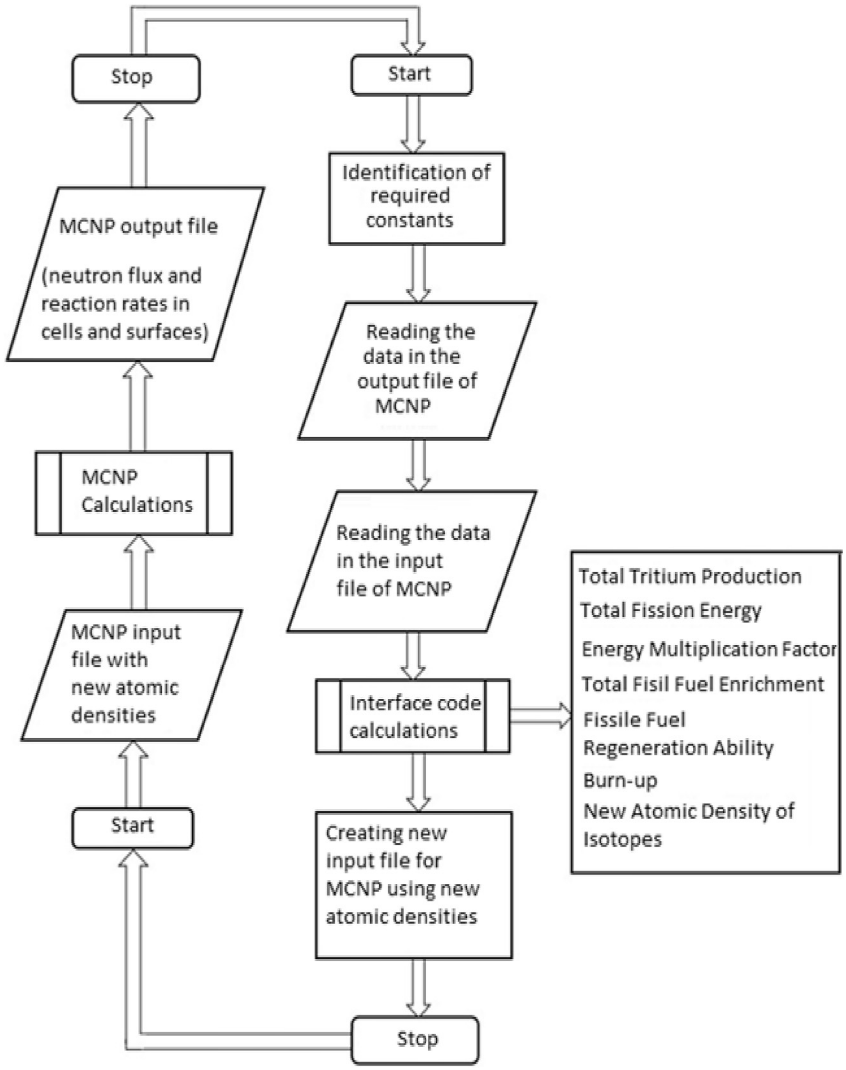

Fig. 1. The flow chart of the interface code.

zones serve as intermediate reflectors and moderators for the escaping fast neutrons from the previous Li_2O zones. The 2nd and 3rd Li_2O zones act as scavengers for the escaping neutrons from previous Li_2O zones for enhanced tritium breeding in ^{6}Li. Two different fissionable fuels have been chosen for the calculations. Firstly, 100% ThO_2 fuel is placed to test the validity of the developed novel interface code. For that purpose, benchmark calculations have been conducted with SCALE code package [3] and ERDEMLI interface code [2]. Afterwards, the same calculations have been performed with MCNP5 1.4v and the novel interface code. Comparison of both results provided the validity of the novel interface code. Secondly, 90% ThO_2 and 10% LWR spent fuel mixture is chosen to perform three-dimensional transmutation and rejuvenation analysis of this fuel. Table- 1 shows the isotopic compositions of the fuels and materials in different zones of the investigated blanket.

3. Numerical results and discussions

As tritium is an artificial element, the tritium breeding ratio of a fusion reactor must be unity for a self-sustaining reactor operation, viz., a fusion reactor must produce more tritium than it consumes.

Fig. 4 shows the variation of the TBR value over operational periods for two different fuel types in the blanket:

- For comparative reasons, the fuel zone of the blanket is first charged with 100% ThO_2 fuel rods, and calculated with the stochastic MCNP5/MCNPAS code set. Afterwards, calculations have been conducted with deterministic SCALE/ERDEMLI set for the same blanket geometry and dimensions using the same ThO_2 fuel rods. In MCNP5/MCNPAS as well as SCALE/ERDEMLI calculations, the blanket remained identical. In the graphic, the red crosses show the TBR values in the blanket, and calculated with SCALE code package using 238 neutron groups' cross sections and the interface ERDEMLI code, whereas the blue circles indicate the TBR values calculated with the MCNP5/MCNPAS code set. One can see in Fig. 4 that the red crosses and blue circles almost overlap each other.
- After setting Benchmark with 100% ThO_2 for the novel code, the fuel zone is charged now with LWR (10%) and ThO_2 (90%) fuel mixture, and calculations are carried out with the MCNP5/MCNPAS codes. In the graphic, the temporal variation of the TBR values for the mixed fuel is shown with the black triangles.

A fusion hybrid blanket operates as fissile fuel breeder, and this increases the neutron multiplication in the blanket, which affects the TBR positively. For both cases TBR increases slightly over the operation time due to a continuous generation and accumulation of

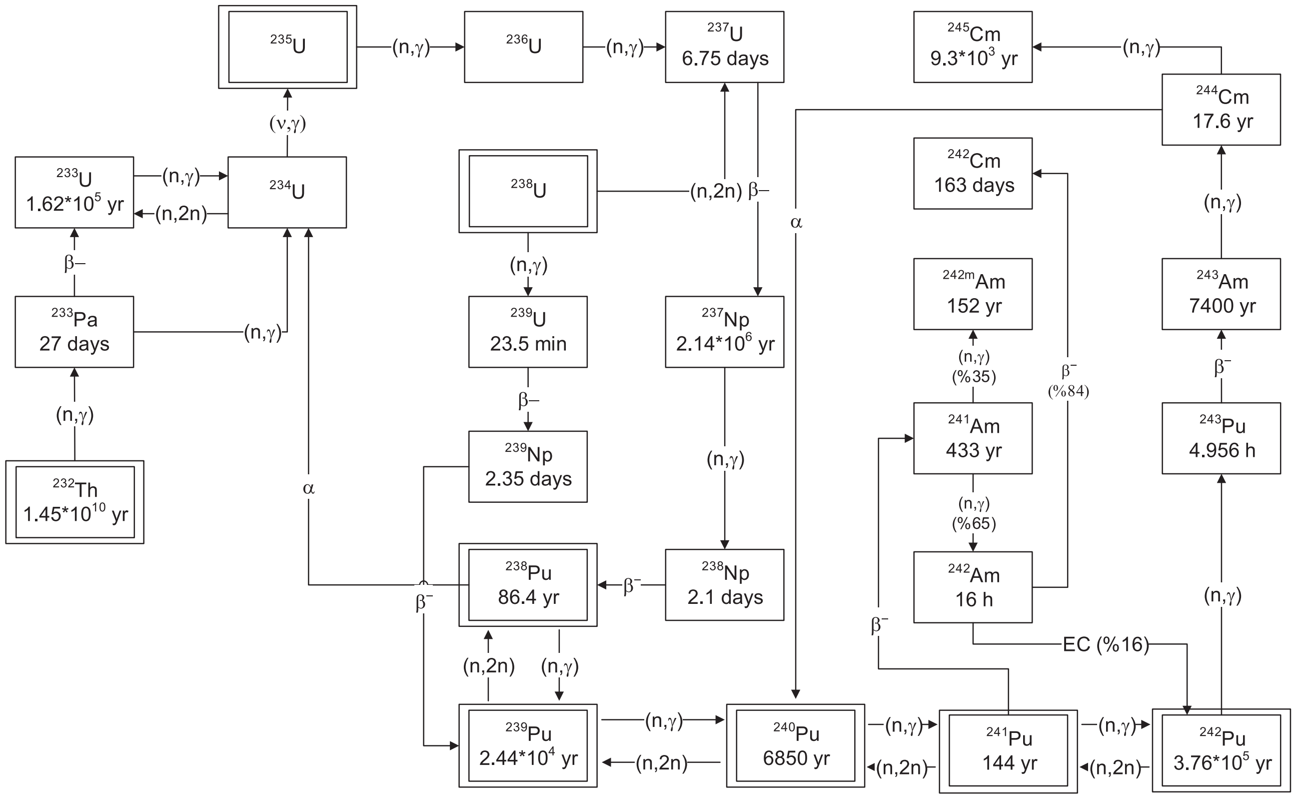

Fig. 2. Transmutation schema of the actinide isotopes.

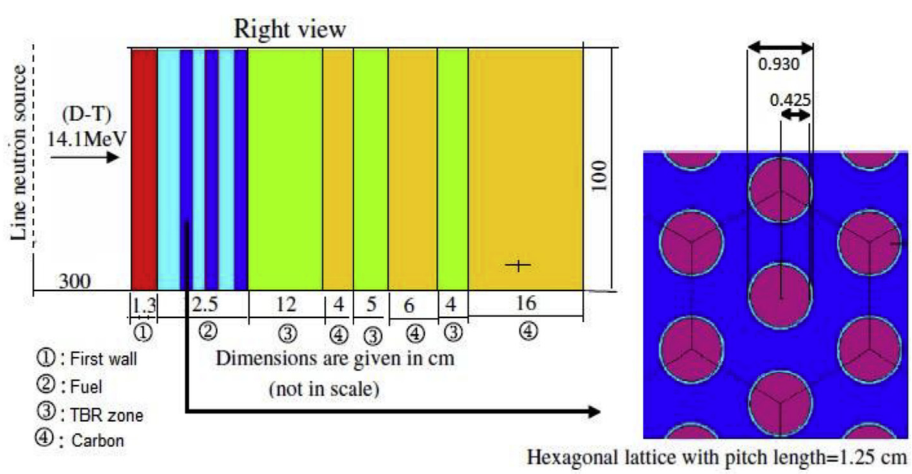

Fig. 3. The cross sectional view of the blanket and fuel region.

^{239}Pu and ^{233}U fuels. The increase of TBR in the blanket with the LWR/ThO_2 fuel mixture is significantly faster than that charged with ThO_2 alone because of the higher neutron multiplication in the LWR spent fuel, especially due to the uranium and plutonium isotopes with higher fission cross sections that thorium.

Excellent agreement of two different code packages verifies clearly the validity of the novel interface code MCNPAS. Minor differences in the results between the two calculations are caused from the fact that SCALE and MCNP codes have different geometry generation methods. Fuel rods are arranged in ten rows in the fuel zone when the geometry is defined for the SCALE code. However, in the MCNP code structure, the program itself forms the geometry of the fuel zone. MNCP places the fuel rods in sequence. When rods come to the edges of the fuel zone, the program creates half or full fuel rods at those points. Thus, there are slight differences in the volume of coolant and the volume of fuel zone compared to the SCALE code. The values of the cross-sections in these libraries may also show minor differences being multi-group for SCALE and continuous for MCNP.

The agreement of the results, calculated with both methods show;

a. The validity of the novel interface code MCNPAS for time evolution calculations in a reactor,
b. The 1-D SCALE code with high energy resolution gives reliable results as the 3-D MCNP code with continuous energy provided

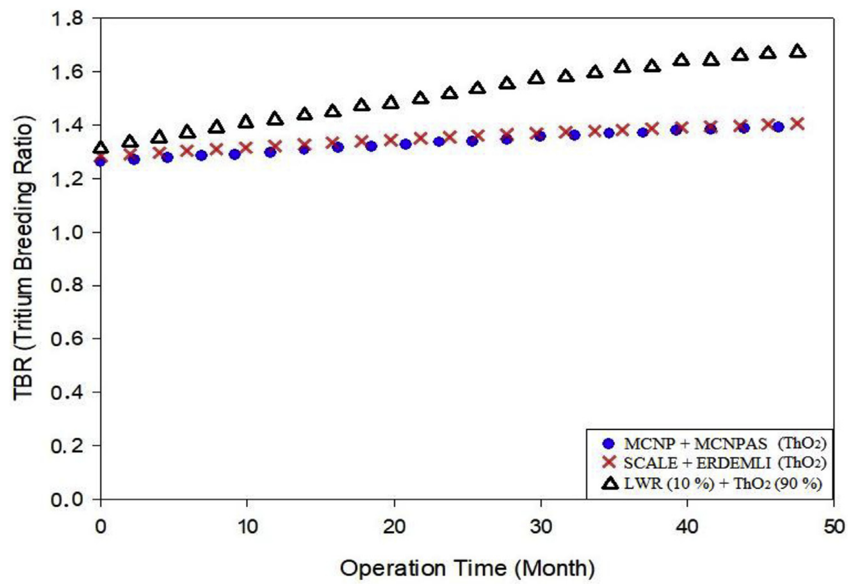

Fig. 4. The change in TBR value during reactor operation time.

that the geometry in not too complex. The former can be preferred for fast generic studies, as the computer run time with SCALE is considerable shorter than the 3-D code MCNP.

In the same graph, the black edge triangular represents the TBR values using 90% ThO_2 and 10% LWR spent fuel mixture, calculated with MCNP5 and MCNPAS.

The threshold fission energy of ^{238}U and ^{232}Th is 1.0 MeV and 1.1 MeV, respectively [49,50]. For 14 MeV neutron energy, the respective fission cross sections become $\sigma_f = 1.2$ b and $\sigma_f = 0.4$ b and the neutron production rates are $\nu = \sim 4$ and $\nu = 3.5$ neutrons per fission. Hence LWR spent fuel and Th act as multipliers in a hybrid blanket under high energetic neutron irradiation [50]. For higher actinides, listed in Table 1, these values are even higher.

Table 1
Atomic densities of the materials in blanket ($10^{24}/cm^3$).

Material	Mass density (g/cm^3)	Zone	Nuclide	Nuclei Density ($10^{24}/cm^3$)
ThO_2	9.86	Fuel	^{232}Th	2.2489E−02
	–	Fuel	^{16}O	4.4978E−02
ThO_2 (90%) + LWR Spent Fuel (10%)	8.00		^{232}Th	2.0239E-02
SS 316L (Stainless Steel)			^{16}O	4.0478E-02
			^{234}U	8.5136E-08
			^{235}U	7.0450E-06
			^{236}U	1.5537E-06
			^{238}U	2.0455E-03
			^{237}Np	3.4054E-07
			^{238}Pu	7.0237E-07
			^{239}Pu	2.5349E-05
			^{240}Pu	2.1944E-05
			^{241}Pu	1.3835E-05
			^{242}Pu	7.8750E-06
			^{241}Am	1.0216E-06
			^{243}Am	2.1922E-06
			^{244}Cm	9.7906E-07
			C	5.6084E-05
			Si	8.7374E-04
			P	4.3496E-05
			S	2.5514E-06
			Cr	1.5084E-02
			Mn	6.1307E-04
			Fe	5.8028E-02
			Ni	1.0575E-02
			Mo	1.1032E-03
Li_2BeF_4	1.94	Coolant (moderator)	^{6}Li	1.7721E-03
			^{7}Li	2.1856E-02
			Be	1.1814E-02
			F	4.7257E-02
Li_2O	2.01	Tritium breeding	^{6}Li	6.0861E−03
			^{7}Li	7.5062E−02
			O	4.0574E−02
Carbon	2.26	Reducing neutron leakage	C	1.1366E-01

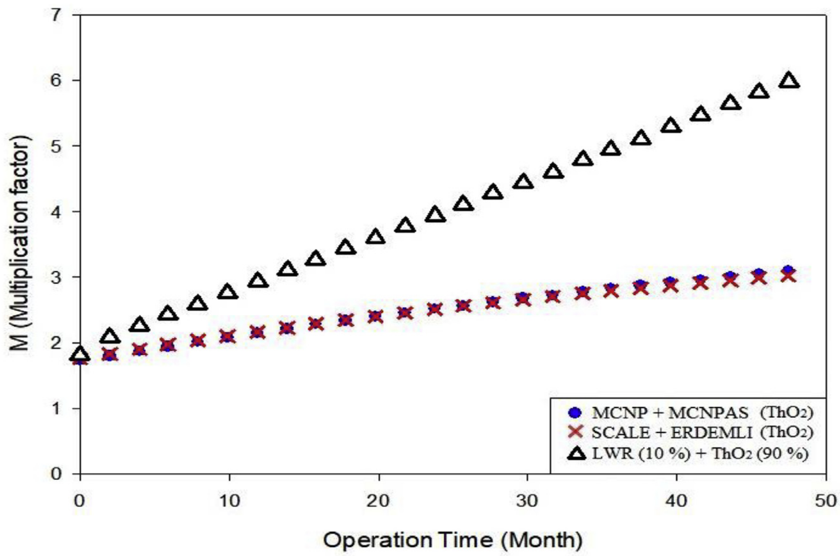

Fig. 5. The change in M value during reactor operation time.

In the course of calculations with MCNP5 and MCNPAS, main neutronic performance parameters of the blanket have been evaluated, accordingly tritium breeding ratio (TBR), multiplication factor (M), cumulative fissile fuel enrichment (CFFE), the net fissile fuel production ability as figure-of-merit (FOM), fissile burn-up, temporal variations and transmutation of actinide isotopes. In terms of showing the continuity of the fusion reaction, TBR value is the most important thing for the reactor operation. TBR value should be greater than 1.05 to sustain deuterium-tritium fusion reaction. As seen in Fig. 4, both fuel types provide the required TBR value. Multiplication factor (M) in Fig. 5 is the ratio between the total energy obtained from blanket through the exoenergetic ^{6}Li(n,α)T and fission reactions and the incident fusion neutron energy. The very small differences in the M values with 100% ThO_2 are due to differences of the modeling of the fuel zone with SCALE and MCNP. One can further see in Fig. 5, higher fission reactions in the uranium and plutonium isotopes in the LWR spent fuel lead to higher energy multiplication than the 100% ThO_2 fuel. The term of the cumulative fissile fuel enrichment (CFFE) is the percentage representation of the fissile fuel produced in the blanket. One can notice the appearance of minor differences between MCNP and SCALE solutions in Fig. 6 over the reactor operation time. In SCALE code calculations, the fuel zone is divided into ten zones and each zone contributes separately to the CFFE value. The CFFE value of the previous region is also seen in the next region in the SCALE calculation so the average value of all regions has been taken for CFFE value. In Fig. 6, red all three lines show the average value in the ten fuel zones. The difference in calculation method of the CFFE value and also minor differences between the cross section libraries of SCALE and MCNP packages as well as different modeling of the fuel zone in both packages cause slight differences. LWRs typically operate with enriched fuel between approximately 3% and 4%. As can be seen in Fig. 6, this value can be reachable in approximately 15–20 months. In particular, 100% ThO_2 fuel can reach this level of

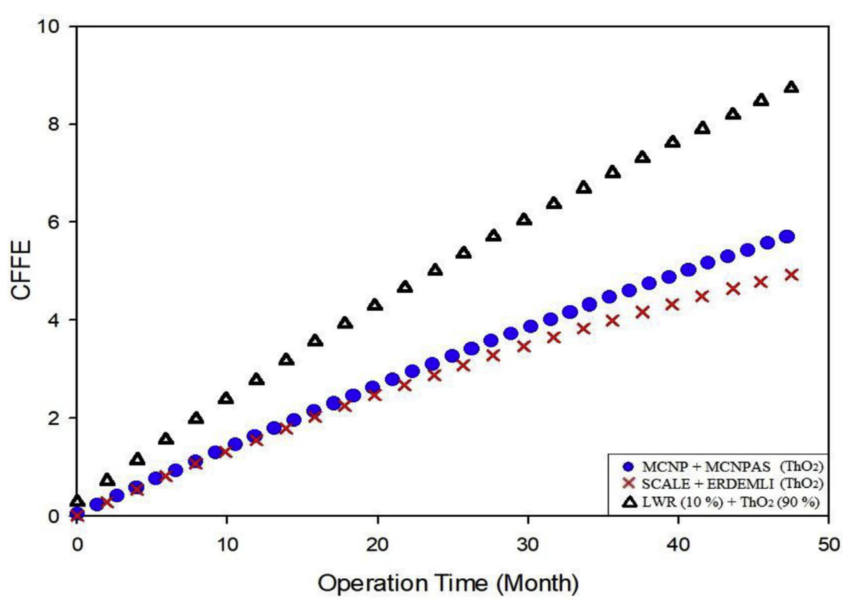

Fig. 6. The change in CFFE value during reactor operation time.

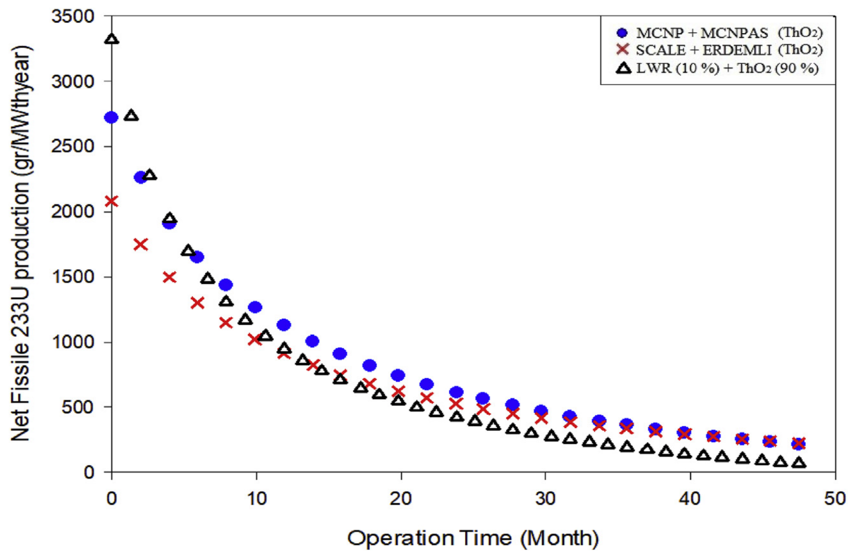

Fig. 7. The net fissile fuel production rate during reactor operation time.

enrichment in about 1.5 years. Furthermore, LWR (10%) and ThO_2 (90%) fuel mixture reach these values at one-year period. This is an important situation; it shows us a fuel that cannot be used directly in the fission reactors and/or thorium by itself can be transformed at longer irradiation in fusion hybrids to usable high quality fissile fuel for reutilization in conventional fission reactors. The ratio between the mass of the fissile isotopes and thermal power of FFHR'S is called the figure-of- merit (FOM). The FOM value is directly related to ^{233}U and ^{239}Pu isotopes. In this study, the production rate of ^{233}U fissile fuel in Fig. 7 is taken as the FOM value. The difference in FOM at the beginning in Fig. 7 arises from the fact that the codes create different volumes when they form the fuel zone. Therefore, the amount of the initial ^{232}Th isotope is different in the modeling of the codes. Continuous production and accumulation of ^{233}U in the course of plant operation increase the fissile fuel burnup and hence suppress the ^{233}U production rate steadily, which is almost disappearing in 48 months. After ~40 months of plant operation, almost all ^{233}U begins to burn *in situ* and the ^{233}U amount nears to saturation. One can observe in Fig. 7 that the LWR spent fuel/ThO_2 curve drops more sharply than the 100% ThO_2 fuel charge case. This is directly related to higher fission isotope content and consequently to higher fission rate in the mixed fuel. MCNP/MCNPAS package calculates higher CFFE values over time than SCALE/ERDEMLI package, which indicates also more drop than the former. This situation indicates that the fission reaction takes place efficiently in the blanket. Burn-up is the measure of the energy obtained from the primary nuclear fuel source. Because of the fuel zone cannot be modeled in the same way at both nuclear codes, initial fuel volume is not same for both codes. Therefore, the burnup values differ for two calculation methods. One can clearly observe in Fig. 8 that LWR spent fuel can deliver about 60 GW.D/MT after four years of operation in a fusion hybrid. The burn-up value of pressurized water reactor (PWR) using 5% enriched uranium is 40 GW.D/MT. In addition, according to literature, with improved fuel assembly designs and fuel management techniques, the fuel burn rate can reach approximately 60 GW.D/MT by increasing the enrichment level above 5% [34]. The LWR spent fuel mixture reaches a slightly higher burn-up rate than this type of PWR reactor. By

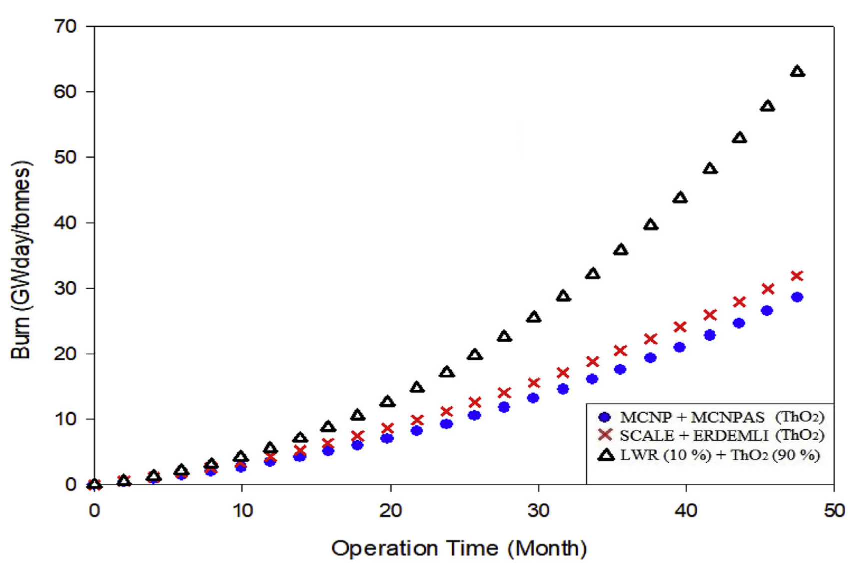

Fig. 8. The fuel burn-up value over the reactor operation time.

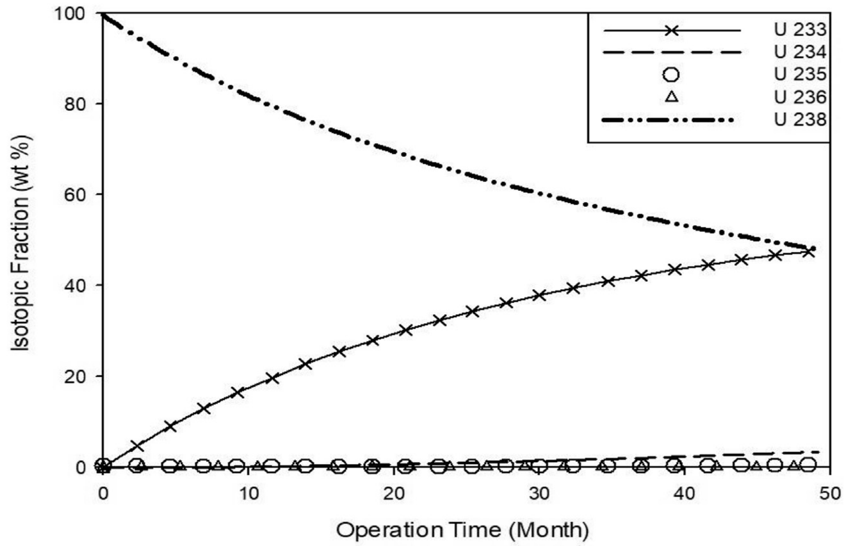

Fig. 9. The change of the isotopic fraction of uranium isotopes in the mixed fuel.

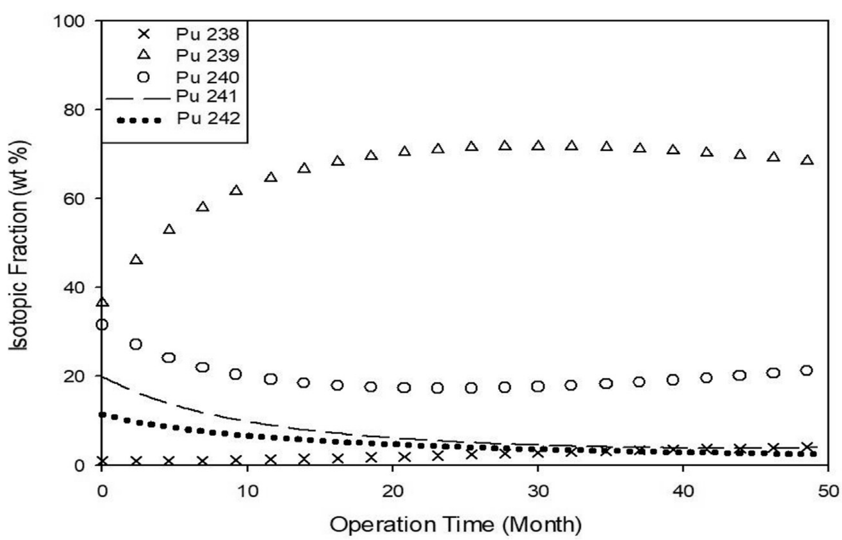

Fig. 10. The change of the isotopic fraction of plutonium isotopes in the mixed fuel.

counting the burn up of fresh fuel in the LWR as well as that of the spent fuel from LWR in a FFHR, it becomes clear that the nuclear waste mass per unit energy drops at least by half by that synergy.

Figs. 9 and 10 depict the isotopic changes in 90% ThO_2 and 10% LWR spent fuel mixture over the plant operation at full power and with a plant factor of 100%. In Fig. 9, one can see the evolution of the uranium isotopes during the four-year operating period. The quantity of ^{238}U isotopes in the LWR spent fuel falls down by half, while the new fissile ^{233}U isotope accumulates up to 45%, which is almost an asymptotic value, as ^{233}U rate becomes nearly negligible by 48 months in Fig. 9. This means medium enriched uranium (MEU) remains at non-prolific level for military applications if separated from the rest of other elements, but it will be highly precious uranium for fast reactors. This has great importance for the reusability of LWR spent fuel after irradiation with (D,T) fusion neutrons. Multiple neutron absorptions in ^{232}Th lead to the production of the fertile ^{234}U isotope, which contributes also to the fissile fuel production through ^{234}U(n,γ) ^{235}U.

Fig. 10 shows the temporal evolution of the plutonium isotopes, which make ~3% of the initial LWR spent fuel, consisting of ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{238}Pu with decreasing fractions. In the first 2 years, ^{239}Pu fraction increases through ^{238}U(n,γ)^{239}Pu. Thereafter, neutron absorption in ^{239}Pu begins to increase ^{240}Pu (n,γ) through ^{239}Pu(n,γ)^{240}Pu. ^{241}Pu and ^{242}Pu falls and ^{238}Pu increases steadily. The high spontaneous fissions in the even plutonium isotopes create such a high level neutron background that makes the plutonium in initial spent fuel fully non-prolific [51]. Neither uranium nor plutonium produced in this blanket can be misused for nuclear weapon production. In addition, the accumulation of fissile plutonium isotopes (^{239}Pu and ^{241}Pu) in the reactor is very favorable in terms of its reuse in LWR reactors.

4. Conclusions

Comparisons for the validity of the created novel interface code showed that the results are generally in a good agreement with previous works on literature. As can be seen in the graphics, there are only slight differences in comparison of the calculations made with SCALE/ERDEMLI and MCNP/MCNPAS code packages for some neutronic parameters. There are diverse reasons causing these differences between SCALE and MCNP solutions. The calculation of the fuel zone volume is carried out in different ways in both codes. In addition, SCALE and MCNP codes solve the neutron transport equation by using different methods. MCNP and SCALE codes have different nuclear data library and neutronic calculations have been performed using these libraries.

The interface codes ERDEMLI and in MCNPAS processes the SCALE and MCNP outputs, respectively, and recalculate the atomic densities of given nuclei after each selected time step in order to create new input files for time-dependent neutronic parameters.

The results further showed that 100 %ThO_2 and its mixture with LWR spent fuel can reach the burn-up value which a LWR reactor reaches during operation. Plutonium component in the LWR spent fuel mixture contains significant fractions of even plutonium isotopes all over the plant operation period and remains at non-prolific level, hence doesn't represent a concern in term of proliferation of nuclear weapons.

Fusion hybrids can make use of thorium fuel with great efficiency. In particular, a mixed fuel composed of thorium and LWR spent fuel can achieve high burn-up rates. Moreover, reusability of LWR spent fuel will have significant contribution to energy production and reduce drastically the nuclear waste of fission reactors.

Acknowledgment

This work is dedicated to the memory of Prof. Dr. Enis ERDİK (1914–1981), University of Ankara, Faculty of Science, Department of Physics, Ankara, TÜRKİYE.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.net.2018.08.006.

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