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Possibility of curium as a fuel for VVER-1200 reactor

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ABSTRACT

In this research, curium oxide (CmO_2) is studied as fuel for VVER-1200 reactor to get an attention to its energy value and possibilities. For this purpose, CmO_2 is used in fuel rods or integrated burnable absorber (IBA) rods with and without UO_2 and then compared with the conventional fuel assembly of VVER-1200 reactor. It is burned to 60 GWd/t by using SRAC-2006 code and JENDL-4.0 data library. From these studies, it is found that CmO_2 is competent like UO_2 as a fuel due to higher fission cross-section of ^{243}Cm and ^{245}Cm isotopes and neutron capture cross-section of ^{244}Cm and ^{246}Cm isotopes. As a result, when some or all of the UO_2 of fuel rods or IBA rods are replaced by CmO_2, we get a similar k-inf like the reference even with lower enrichment UO_2 fuels. These studies show that the use of CmO_2 as IBA rods is more effective than the fuel rods considering the initially loaded amount, power peaking factor (PPF), fuel temperature and void coefficient, and the quality of spent fuel. From a detailed study, 3% CmO_2 with inert material ZrO_2 in IBA rods are recommended for the VVER-1200 reactor assembly from the once through concept.

1. Introduction

Curium is a transuranic radioactive element of the actinide series. Curium is present in wastes resulting from the reprocessing of the irradiated fuel of nuclear power reactors. Consequently, a large potential supply will be available in future years. Estimates indicate that its cost will be substantially lower than that of ^{238}Pu [1]. Most curium is produced in the nuclear reactors from the bombardment of uranium and plutonium with neutrons. So an amount of curium is present in the spent nuclear fuel, it's roughly about 64.7g/ton after 60 GWd/t burnup for a 4.73% enriched LEU fuel [2]. Almost all isotopes of curium from ^{242}Cm and ^{248}Cm, as well as ^{250}Cm, have a significant fission cross-section and undergo a self-sustaining nuclear chain reaction. Like most of the transuranic elements, the fission cross-section of the odd-mass curium isotopes ^{243}Cm, ^{245}Cm and ^{247}Cm are higher than the even-mass curium isotopes. For having large fission cross-section, it can be said that curium can also be used as a fuel like the conventional fissile elements for any thermonuclear reactors.

The fission properties of curium separated from spent nuclear fuel have been studied [2]. A neutronic study for the possibility of burning curium as a fuel has been done for a fast reactor [3]. For understanding the transmutation behavior of curium in MOX Fuel, it has been irradiated in the experimental fast reactor JOYO [4]. Different aspects of the fabrication of curium-based fuels have been studied [5].

Due to the low availability and high price of curium, till now it has not been used as a nuclear fuel. But the stockpile of curium is increasing continuously as it is generating from the nuclear reactors. Then it is separated from the spent fuel as it is a great source of decay heat. Even more curium is generated now from the burning of plutonium as many countries are ongoing with the projects of reduction of weapon-grade plutonium stockpile. So, soon there will an increase in curium stockpile. As ^{245}Cm and ^{247}Cm have very high fission cross-section, small critical masses and very high half-life, therefore it could be used in nuclear weapons instead of conventional ^{239}Pu. In this situation, for proliferation resistance, we need to burn the curium for reducing its stockpile.

The VVER-1200 reactor is the very first nuclear reactor in our country (Bangladesh) and it is under construction at Rooppur Nuclear Power Plant (RNPP) facility. So, we took a project for improving and understanding our upcoming reactor in various aspects. As a part of this project, earlier we studied the possibility of americium as a burnable absorber for VVER-1200 reactor [6]. In this

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project, we studied the possibility of curium oxide (CmO_2) as a fuel for the VVER-1200 reactor and detailed neutronic characteristics have been studied. For this purpose, we have considered different percentages of CmO_2 and mixed homogenously with all the fuel rods as well as only in the integral burnable absorber (IBA) rods in the assembly. Then neutronic calculations have been conducted and compared with the reference assembly for finding out the (a) feasibility of using curium as a fuel, (b) fuel enrichment reduction impact and (c) use of curium as a once through fuel for the VVER-1200 fuel assembly considering the infinite multiplication factor (k-inf), power peaking factor (PPF), safety factors and quality of spent fuel.

1.1. Materials and method

The VVER-1200 reactor assembly contains a total of 331 rods where 312 are fuel rods and others are central tube and control rod guide tubes. Among the fuel rods, 12 rods are homogenized with UO_2 and Gd_2O_3 as integrated burnable absorber (IBA) rods. A horizontal cross-sectional view of the reference assembly and a cell of that assembly are given in Fig. 1. The parameters considered for modeling the reference assembly are given in Table 1. The cosidered models for this analysis are given in Table 2. The neutronic calculation has been carried out with modeling by SRAC-2006 code and JENDL-4.0 nuclear data library [7,8]. SRAC (Standard Reactor Analysis Code)-2006 is a deterministic code for nuclear reactor design and analysis developed by Japan Atomic Energy Agency (JAEA). The PIJ module has been used here, which is a collision probability method (CPM), and the geometry used is a hexagonal assembly of 60° rotational symmetry with pin rods on a triangular mesh. One-sixth of the assembly image is given as the code considers it as a 1/6th symmetrical assembly and the output of the geometrical model of the designed assembly are given in Fig. 2. The composition of *Cm* is used from the spent fuels of VVER-1200 assembly with 4.95-wt% enriched UO_2 fuel after 60 GWd/t discharge burnup and 10 years cooling. The composition of *Cm* isotopes $^{243}Cm/^{244}Cm/^{245}Cm/^{246}Cm$ is 0.78/90.07/8.08/1.07 wt% respectively with a density of 13.51 g/cm^3.

2. Infinite multiplication factor (K-inf)

The variation of k-inf with burnup for the different investigated fuel models in comparison with reference are shown in Fig. 3 (a, b, c, d, and e). Fig. 3 (a) illustrates the k-inf values for the case of homogenized mixer Model-1 (CmO_2 is blended with UO_2 and used as a fuel rod). K-inf is almost similar to the reference throughout the burnup period. The thermal fission cross-section of ^{243}Cm, ^{244}Cm, ^{245}Cm and ^{246}Cm isotopes are 587.4 b, 1.022 b, 2054 b and 0.44 b respectively [7]. ^{243}Cm has an almost similar fission cross-section to ^{235}U (585.1 b). The ^{245}Cm is also a great fissile element as it has a high fission cross-section of 2054 b. The major constituent of these *Cm* isotopes is ^{244}Cm which is a fertile element. Almost 81% of ^{244}Cm converts to ^{245}Cm which is a great fissile element [9]. So if we use *Cm* in fuel, the fission rate increases and the k-inf rise. Due to this reason with increasing concentration of *Cm*, higher k-inf is gained. A homogenized mixer of 0.2% CmO_2 with UO_2 in fuel rods shows the most compatible k-inf. The amount of curium needed in this fuel design is 0.0187g/cc.

The homogenization of *Cm* in all fuel rods is quite difficult. So the use of *Cm* in only the IBA rods has been studied here. Fig. 3 (b) shows the k-inf values with respect to the reference one for the case of *Cm* homogenized mixer only in the integral burnable absorber (IBA) rods Model-2 (CmO_2 is blended with UO_2 and used in IBA rods). In this type different percentage of *Cm* is taken for analysis. The 3% CmO_2 in the IBA rods shows exactly the same k-inf trend as the reference assembly. The amount of curium needed in the IBA rods is 0.27 g/cc. The higher percentage of *Cm* in IBA rods leads to higher k-inf values as *Cm* acts like uranium fuel with having both fissile and fertile nuclides.

As the curium contains fissile ^{243}Cm and ^{245}Cm, we tried to replace the 3.6 wt% enriched uranium with natural uranium to investigate the effect. Fig. 3 (c) shows the k-inf values with respect to the reference for the case of *Cm* homogenized mixer only in the

Table 1
Design parameters considered for the reference VVER-1200 fuel assembly.

Parameters	Unit	Value
Fuel pellet hole radius	mm	0.60
Fuel pellet outer radius	mm	3.80
Pin pitch	mm	12.75
Fuel temperature	K	873.00
Non-fuel temperature	K	573.00
^{235}U enrichment in fuel rods	wt%	4.95
Moderator		Light water
Boron concentration in moderator	ppm	400

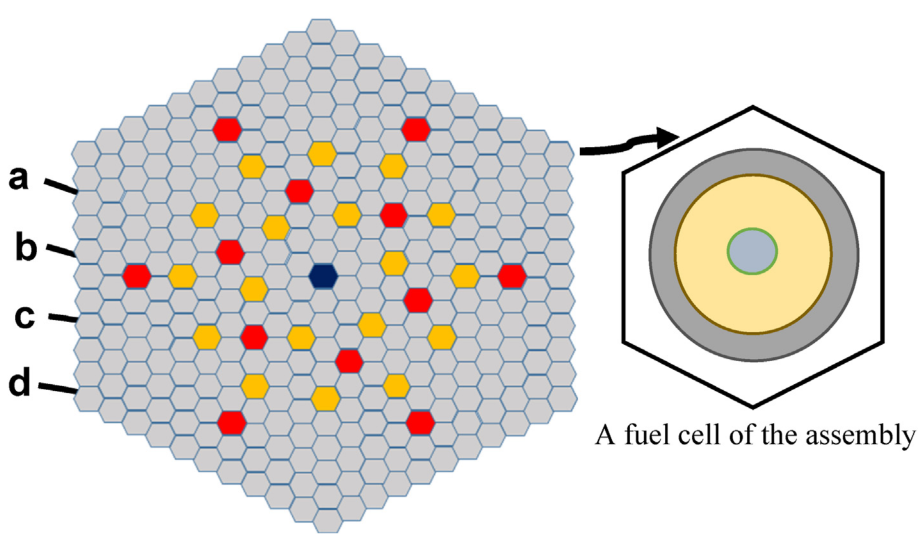


Fig. 1. Horizontal cross-section of reference fuel assembly of VVER-1200 reactor (model Ref): (a) central tube; (b) control rod guide tubes; (c) IBA fuel rod (3.6% ^{235}U +4% Gd_2O_3); (d) fuel rod (4.95% enriched UO_2).

Table 2
Considered models for analysis.

Model Name	Fuel Rod (All percentages are in wt%)	IBA Rod (All percentages are in wt%)
Reference	UO_2 [En-4.95%]	UO_2 [En 3.6%] + Gd_2O_3 (4%)
Model-1	Homogeneous mixture of UO_2 [En-4.95%]+ CmO_2 (0.1%, 0.2%, 0.3%,0.4%, 0.5%,respectively)	UO_2 [En 3.6%] + Gd_2O_3 (4%)
Model-2	UO_2 [En-4.95%]	UO_2 [En 3.6%] + CmO_2 (1%, 2%, 3%, 5%, 7%, respectively) + Gd_2O_3 (4%)
Model-3	UO_2 [En-4.95%]	UO_2 [Nat-U] + CmO_2 (1%, 3%, 5%, 7%, 10%, 12%, respectively) + Gd_2O_3 (4%)
Model-4	UO_2 [En-4.95%]	CmO_2 + Gd_2O_3 (4%) CmO_2 + Gd_2O_3 (4%) [volume is reduced by 60%] CmO_2 + Gd_2O_3 (4%) [volume is reduced by 70%] CmO_2 + Gd_2O_3 (4%)[volume is reduced by 80%]
Model-5	UO_2 [En-4.95%]	ZrO_2 + CmO_2 (1%, 3%, 5%, 7%, 10%, 12%, 15%, respectively) + Gd_2O_3 (4%)

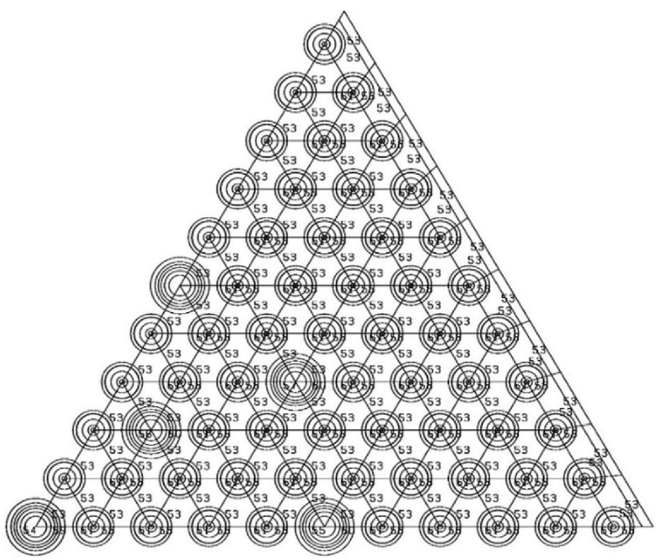


Fig. 2. Geometrical image output of 1/6th of the VVER-1200 fuel assembly generated from SRAC-2006 code.

integral burnable absorber (IBA) rods, Model-3 (CmO_2 is blended with natural/depleted UO_2 and used in IBA rods). In this type different percentage of *Cm* is taken for analysis and 3% CmO_2 with natural Uranium in IBA rods shows better k-inf with respect to the reference model. As the IBA rods are made with natural uranium, the cost of fuel enrichment and fabrication is reduced here. So, it is a promising model and can be studied further.

From the above discussion, it can be said that curium can be used as fuel like uranium and the use of it in IBA rods show good performance. So here we replaced the uranium of IBA rods with curium in Model-4. As curium contains a high amount of fissile element the k-inf increases largely. To reduce the effect, the IBA rods are shrunk in volume to reduce the quantity of curium. In Fig. 3 (d) a comparison of k-inf with the reference model, actual volume IBA rods loaded with *Cm* and IBA rods with reduced volume is shown. The 20% of the original volume shows a slightly better percentage than the other models apparently. But if we use the actual volume IBA rods, there is a scope for enrichment reduction of the whole assembly. The enrichment can be reduced to 3.5 wt% which is almost a 30% reduction in enrichment. Fig. 4 (a) illustrates that the k-inf curve with reduced enrichment of 3.5 wt% shows exactly the same as the reference assembly. But in this model, a higher amount of *Cm* (10.78g/cc) is needed. To solve this problem reduced volume IBA rods can be used. Fig. 4 (b) shows the k-inf curve of 80% reduced volume of IBA with lower enrichment of the whole assembly. The enrichment can be lowered up to 4.5 wt% but the excess reactivity is higher at the BOL. If we use 8% Gd_2O_3 instead of 4% in the IBA rods, it suppresses the higher excess reactivity and shows a good k-inf trend comparing with the reference assembly.

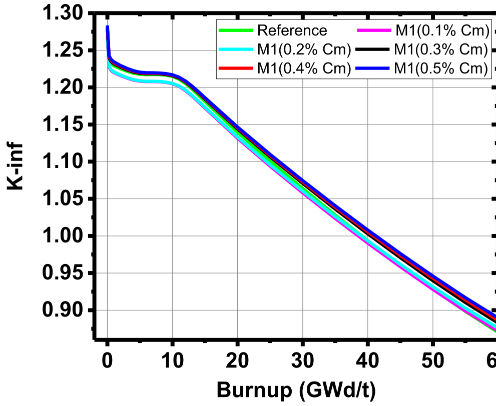
As a high quantity of curium is needed in Model-4, a new idea is proposed here to use inert matrix elements in the IBA rods. Zirconia (ZrO_2) is used as an inert matrix element. Fig. 3 (e) shows the k-inf variation for Model-5 (inert matrix fuel or once-through zirconia fuel). In this model, the IBA rods are homogenized with only Zirconia, CmO_2 and Gadolinia. Different percentage mixtures of zirconia and CmO_2 are taken for analysis. Among these models, 91% ZrO_2 + 3% CmO_2 + 4% Gd_2O_3 shows better k-inf with respect to the reference models and also the amount of curium needed in the IBA rods is 0.27 g/cc which is lower than other prospective models. Though there is higher excess reactivity in the BOL than the reference model, it can be suppressed by adding 8% Gd_2O_3 instead of 4% Gd_2O_3. Fig. 4(c) shows that adding additional Gd_2O_3 in the IBA rods makes the burnup dependent k-inf trend exactly the same as the reference model. Due to the low solubility of the inert matrix fuel, it can be easily disposed through geological disposal. So, it is a very attractive option in terms of spent nuclear fuel (SNF) management. These IBA rods can also be used as once-through fuel rods which can be further investigated.

3. Power peaking factor (PPF)

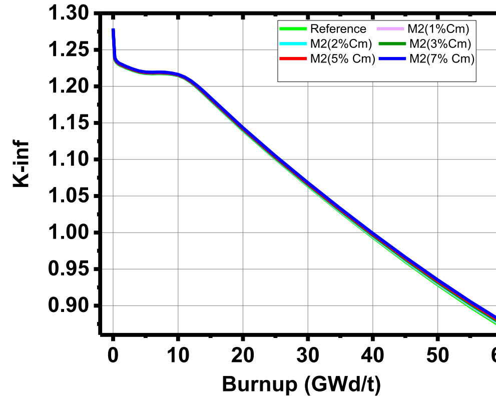
From the k-inf analysis, several potential models have been analyzed and preliminarily found to be a good option. But for choosing the best model, we have to analyze these models considering the power peaking factor (PPF). From the k-inf figures, it is seen that 0.20% of Model-1, 3% of Model-2, Model-3, 80% shrink of Model-4, and 3% of Model-5 are the potential models and further analysis have been done for these models.

The power peaking factor is an important operational and safety parameter for any PWR. As it is the ratio of the maximum pin power to the average power of the assembly, the goal is to reduce PPF as much as possible. A limit for PPF is set for each type of reactors according to the design and different safety limits. Higher PPF indicates that there are some fuel rods with higher temperatures than other rods and if this value crosses the safety limit, the heated fuel rods may cross the safe temperature limit and may cause fuel rod oxidation and even fuel clad rupture or even meltdown. So, the goal is to minimize the value of PPF as much as possible.

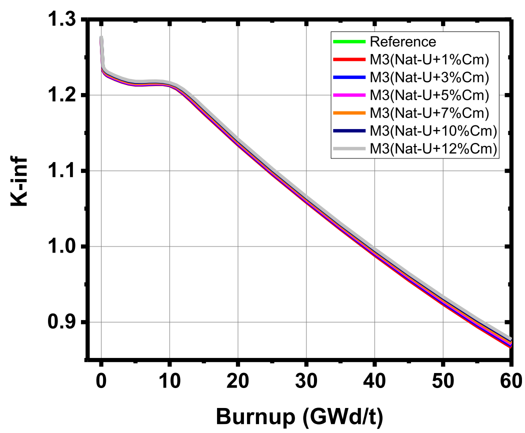
The PPF graph of the potential designs is shown in Fig. 5 (a). Model-1 shows a very high PPF than the other models just after the startup. So, for better understanding the differences among other models PPFs, it has been eliminated. From this figure, it is seen that



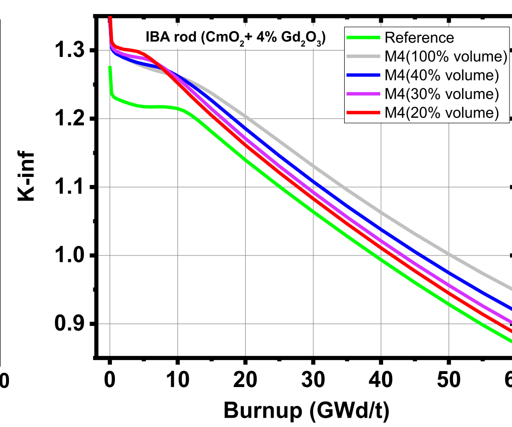
a. Cm mixed with UO_2in FRs



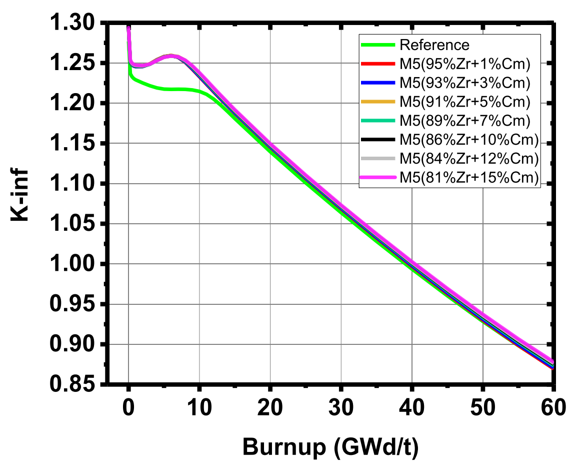
b. Cm mixed with UO_2in IBA rods



c. Cm mixed with Nat-UO_2 in IBA rods



d. Only Cm is used in IBA rods



e. Cm mixed with ZrO_2 in IBA rods

Fig. 3. Burnup dependent k-inf for different Models in comparison with reference.

Model-4 shows the best result in terms of PPF but considering the k-inf and initial loading weight of *Cm*, it is the least attractive model. On the other hand, Model-5 is the most attractive model in all these aspects as it has lower IBA rod power and shows less fluctuation in PPF with respect to burnup. Though it has a slightly higher PPF factor than other models, the difference is very low and the reason for this slightly higher PPF is the lower power in the IBA rods than the other fuel rods. As the IBA rods contain gadolinia which has less heat conductivity than UO_2. Therefore, higher heat generation in the IBA rods might cause the melt down due to its

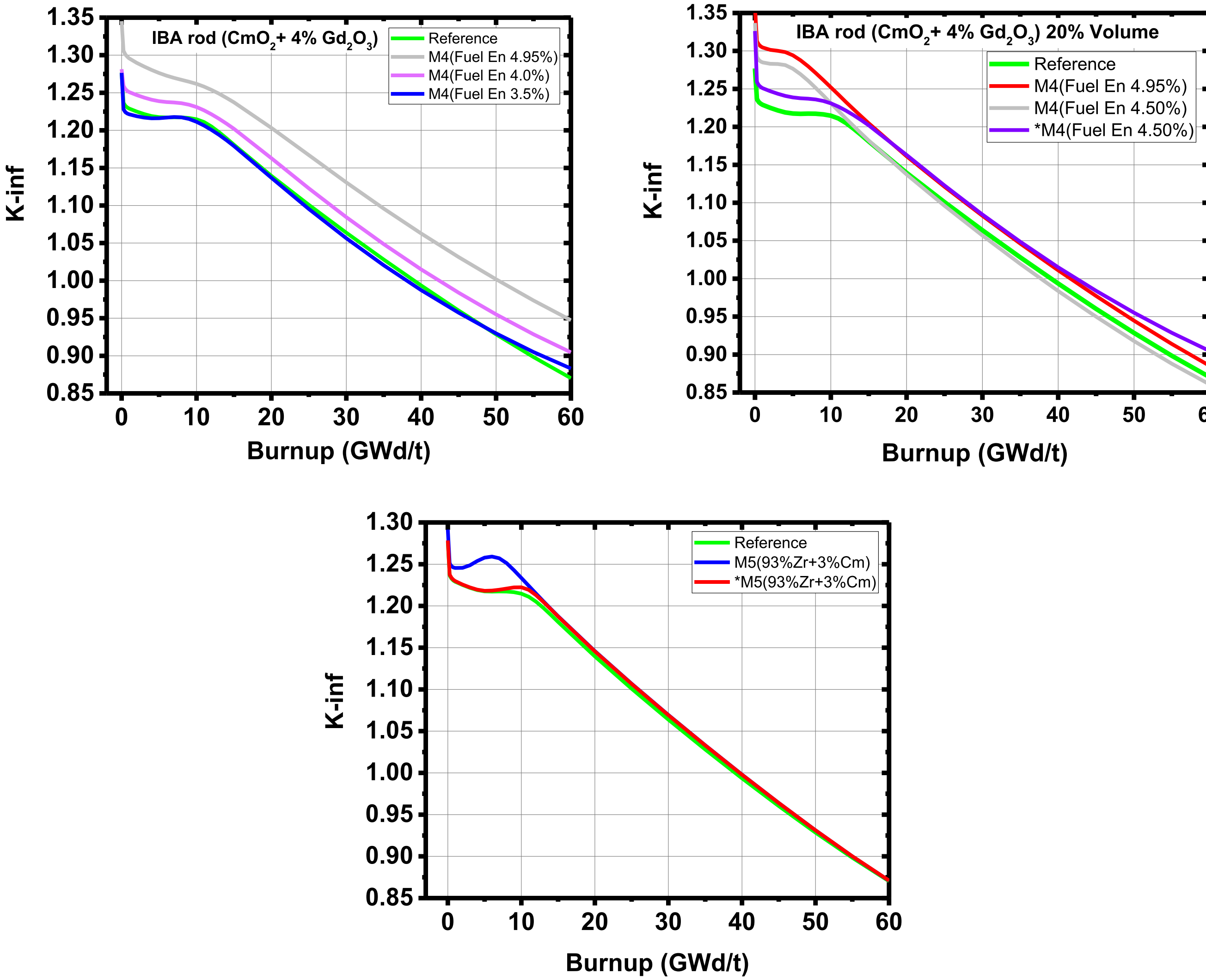
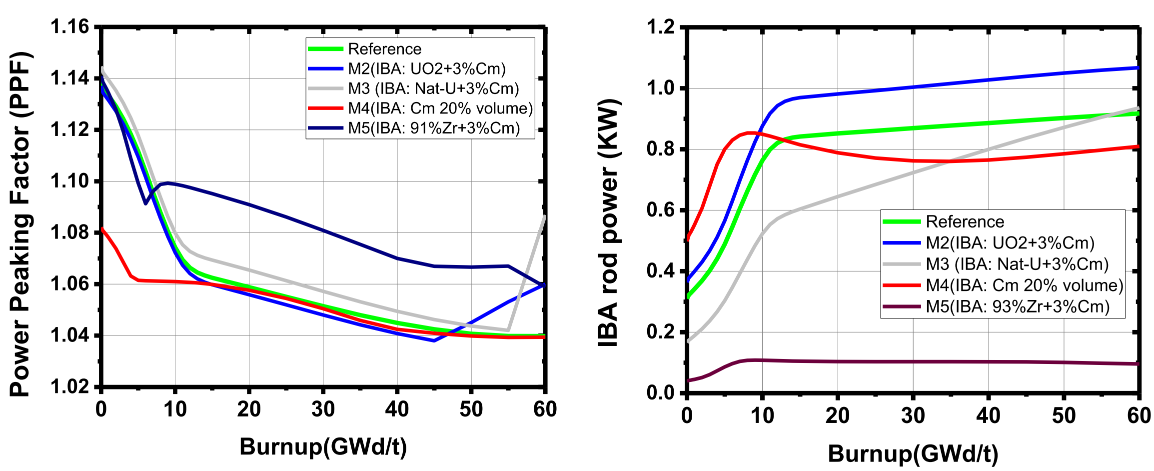


Fig. 4. (a). Burnup dependent k-inf for Model-4 with reduced enrichment with actual volume of IBA rods
Fig. 4 (b). Burnup dependent k-inf for Model-4 with reduced enrichment with 20% volume of IBA rods. [*M4 (8% Gd_2O_3 is used to decrease the excess reactivity)]
Fig. 4 (c) Burnup dependent k-inf for Model-5 with 8% Gd_2O_3.



(a) (b)

Fig. 5. Burnup dependent (a) power peaking factor (b) IBA rod power of the potential models and the reference fuel assembly.

lower heat transfer. So, the heat generation in IBA rods has to be lower than the fuel rods. Fig. 5 (b) shows the IBA rod average pin power, it is lower than the average pin power of the assembly as the IBA rods are uranium free and the *Cm* is only 3 wt%. Due to this lower pin power of the IBA rods and this power difference between the IBA rod and other pin rods, the PPF becomes slightly higher than the other models. Moreover in this model, the IBA rods are used as once-through fuel so it would be a good option for proliferation resistance and SNF management. Further analysis would be continued for this model. Model-2 and Model-3 also show similar lower PPF but after 45 GWd/t burnup, the PPF increases drastically due to the buildup of new fissile isotopes from the fertile ones, especially in Model-3.

4. Safety factors

The fuel temperature coefficient (FTC) and void coefficient (VC) are the basic thermal safety parameters of any nuclear reactors. For the stable power operation of any reactor, FTC and VC must be negative. With negative coefficients, when fuel or moderator temperature increases, density decreases, which decreases the resonance escape probability as well as increases the thermal utilization factor.

4.1. Fuel temperature coefficient (FTC)

The fuel temperature coefficient (FTC) has been calculated for the potential models when the fuel temperature is increased by 300 K. Fig. 6 shows the burnup dependent fuel temperature coefficient (FTC) for the potential models. As we know, with increasing the temperature, the relative thermal motion of fuel atoms and neutrons increases which broadens the resonances capture cross-section of ^{238}U especially at the beginning of life (BOL) and provides strong negative reactivity with increasing the fuel temperature. For these reason, the FTC of potential models shows more negative at the BOL. But, fissile ^{239}Pu is buildup from ^{238}U with burnup in these fuels and contributes an increasingly positive component to FTC. At the end of life (EOL) FTC of Model-3 becomes more positive than the remaining used models. This is because; in Model-3 natural uranium is used instead of 3.6% enriched uranium, which increases the production of ^{239}Pu. However, it is expected that the mean core FTC in VVER-1200 reactor will be negative over reactor life because spent fuel is continuously replaced by refueling with burnt-up of fuel.

4.2. Void coefficient (VC)

When the reactor coolant temperature is increased, the density decreases. For the water-cooled reactors, boiling and void take place. As the density of steam is lesser than that of water, the production of steam can be considered as an equivalent of producing voids in the water. We have increased the void fraction from 0% to 40%, for calculating the void coefficient. This caused a reduction in the amount of coolant and also the scattering, moderation, and capture. This resulted in the absorption of fewer neutrons by the fuel. So, the reactivity decreases consequently. Fig. 7 shows the void coefficients of the potential models in comparison with the reference.

5. Quality of spent fuels (QSF)

In this section, spent fuel is analysis for the Model-5 in comparison with the reference. In the discharged fuel of Model-5, 44.34% of initially loaded *Cm* has been transmuted excluding the *Cm* produced in the reference model. Fig. 8 (a) illustrates the burnup dependent inventory of *Cm* for Model-5 and the reference fuel assembly. As *Cm* is initially loaded in Model-5 the quantity is higher than the reference. The trend of plutonium isotopes throughout the burnup period of Model-5 with respect to the reference assembly is shown in Fig. 8 (b). In Model-5, the production of Pu is 5.35% less than the reference. It is expected that the U-free IBA rods increases the fission of Pu in Model-5. Fig. 8(c) illustrates that the production of Am isotopes in Model-5, which is 5.80% less than the reference assembly. Moreover, the production of Np is 4.44% less than the reference. The production of LLFPs is shown in Fig. 8(d). From this figure, it can be observed that Model-5 has lower production of LLFPs than the reference assembly after the burnup period of 60 GWd/t. This is because ^{243}Cm and ^{245}Cm have a lower fission yield of LLFPs than the ^{235}U isotope. By the analysis of discharged IBA rods, it is found that 41.49% of the initially loaded curium is transmuted in which, 3.69% of the *Cm* is converted into Pu and 0.48% is converted into Am.

6. Conclusions

The objective of this research is to find out the possibility of curium oxide (CmO_2) as a fuel for VVER-1200 reactor by using it as a homogeneous mixture with UO_2 and individually as fuel rods or integrated burnable absorber (IBA) rods. For this purpose, detailed

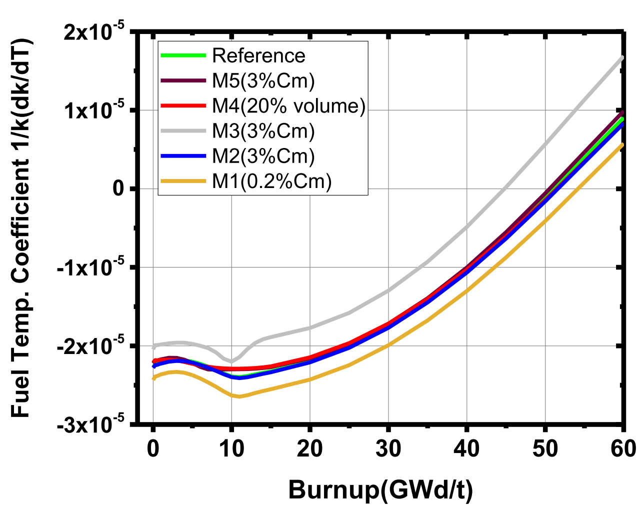


Fig. 6. Burnup dependent fuel temperature coefficient for potential models.

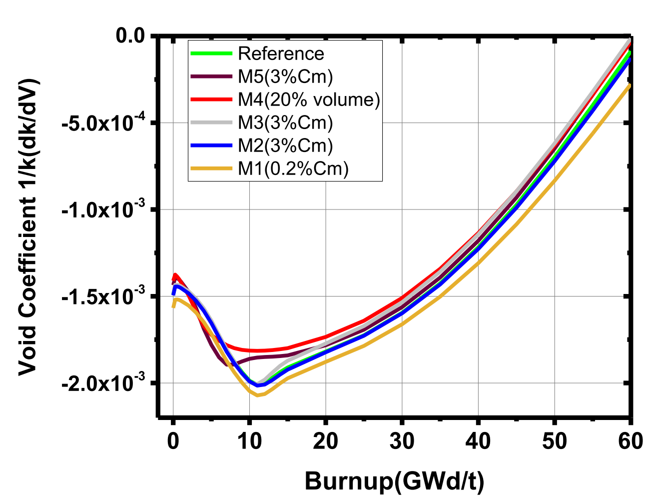


Fig. 7. Burnup dependent void coefficient for potential models.

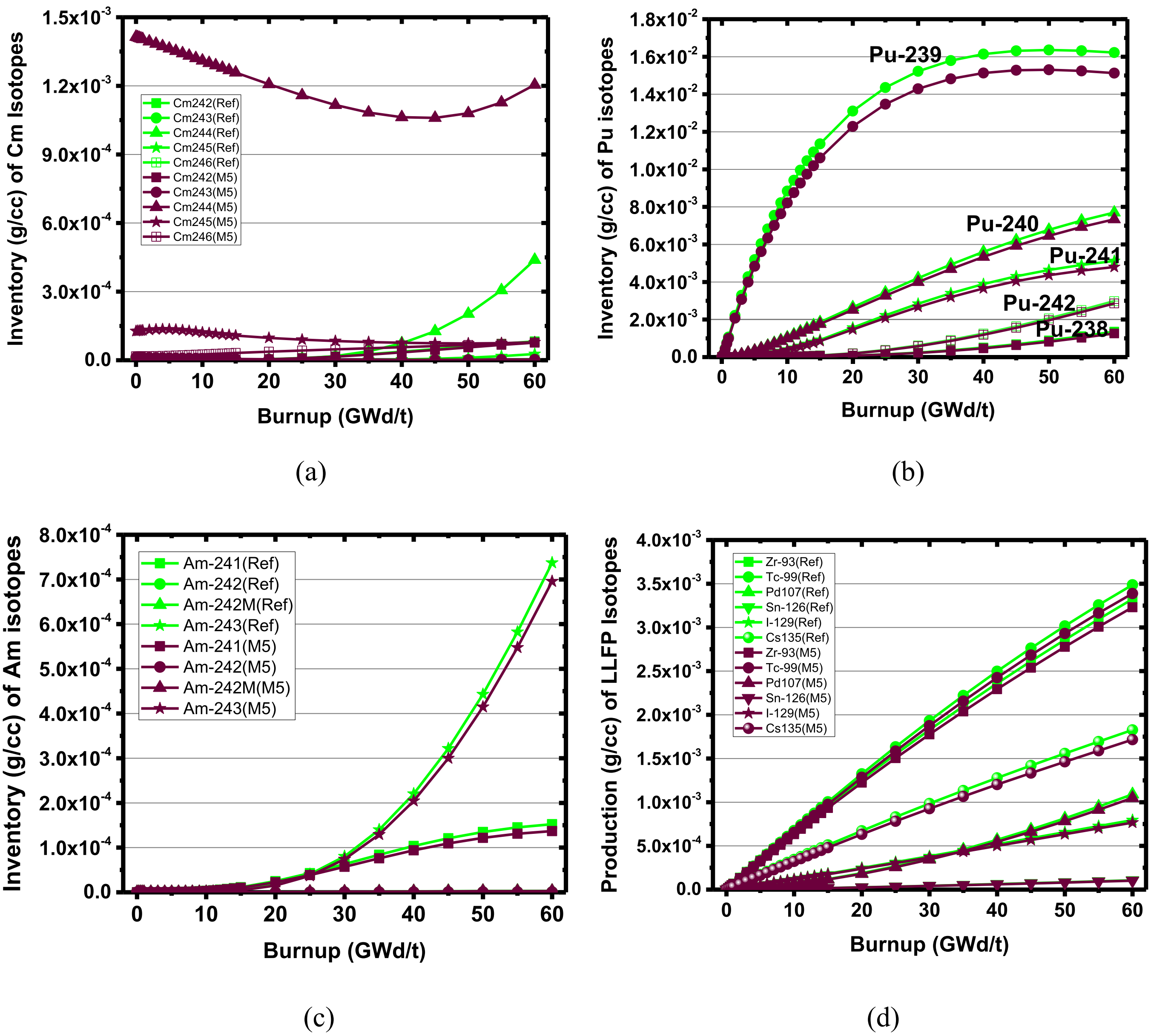


Fig. 8. Burnup dependent inventory of (a) *Cm*, (b) Pu, (c) Am, and (d) LLFPs of the Model-5 and the reference assembly.

neutronic characteristics are studied to find its energy value and compared with the reference assembly considering the infinite multiplication factor (k-inf), power peaking factor (PPF), safety factors and quality of spent fuel. The effect of fuel enrichment reduction impact and use of curium as a once through fuel are also studied. It is found from the k-inf that curium can be used as fuel like uranium and the potential models are 0.20% CmO_2 for Model-1, 3% CmO_2 of Model-2, 3% for Model-3, 80% shrunk volume for Model-4, and 3% for Model-5. The use of CmO_2 in IBA rods is more effective than using in the fuel rods. In the studied models, Model-5 (93%ZrO_2+3%CmO_2+4%Gd_2O_3) is recommended after considering its safety parameters and spent fuel quality. In this model, 0.27 g/cc of CmO_2 is needed initially, but the discharged amount of *Cm* is 41.49% less than the initial. In the discharged IBA rod, the amount of Pu is only 3.69% of initially loaded *Cm* and Am is 0.48%. Therefore, the quality of discharged IBA rods will be less attractive concerning to proliferation. This is because of the absence of uranium and the presence of inert material (ZrO_2) which could be proposed as a once through use.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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