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Article

Comparative Assessment of Molten Salt Reactor Neutronic Performance with Various U-233 Purity

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Abstract. Molten salt reactor (MSR) can be deployed as a thermal breeder reactor in a thorium fuel cycle. The fissile nuclide mostly uses U-233, which is nonexistent in nature and must be synthetised. Researches on thermal breeder MSR usually assume that the U-233 is pure, but in technical reality, U-233 synthesis always accompanied by other uranium isotopes. These impurities can affect the reactor physics performance and altering the operational safety consideration. This research studies the impact of using impure U-233 on the neutronic performance Passive Compact Molten Salt Reactor (PCMSR). Four U-233 vectors with various level of purity were used as comparison. The investigated parameters were reactor criticality, temperature coefficient of reactivity (TCR), kinetic parameters, and conversion ratio (CR). The calculations were performed using MCNP6.2 code with ENDF/B-VII.0 nuclear data library. From the calculation, impure U-233 fuels were proved to improve the TCR as a result of weaker moderator temperature coefficient (MTC). Whilst impurity does not particularly affect delayed neutron fraction, it reduces neutron generation time. Impure U-233 vectors slightly altered CR value, but rather insignificant. Overall, operational safety and CR value can be maintained even if the MSR core is started using low-purity U-233.

Keywords: PCMSR, U-233, MCNP6.2, ENDF/B-VII.0.

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1. Introduction

The resurgence of interest on molten salt reactor (MSR) as a candidate of Generation IV (GenIV) nuclear reactor technology drove its development in recent decades. Compared to conventional light water reactors (LWRs), MSR possesses several advantages, such as low operating pressure, high operating temperature, online fission product removal, and the use of chemically stable salt compound to prevent unwanted release of fission product to the environment. The heat from its high operating temperature can be used to produce hydrogen. Provided that proper neutron economy can be achieved, MSR can also operate as a thermal breeder reactor using thorium fuel cycle. This is further supported by the possibility of online fuel reprocessing inherent to MSR, allowing the Pa-233 precursor to be removed from core, decayed into U-233, and then re-injected into the core [1]–[4].

Thermal breeder MSR was previously developed in Oak Ridge National Laboratory (ORNL) in the form of Molten Salt Breeder Reactor (MSBR). However, before the design can be realised even as a prototype, the development was abruptly halted in the 1970's [3], [5]. In Japan, MSR-FUJI was developed as a self-sustaining MSR without online reprocessing system [6], [7]. Currently, several thermal breeder MSRs are being developed by various entities, such as LFTR (Flibe Energy, USA) [8], SD-TMSR (SINAP, China) [9], and PCMSR (UGM, Indonesia) [10]–[12].

The main premise of an MSR is its liquid fuel, which act simultaneously as the coolant. Uranium and thorium, in a fluoride compound is dissolved within a eutectic carrier salt, typically lithium fluoride (LiF) and beryllium fluoride (BeF_2). The salt is melted into liquid form at high temperature (> 600°C), then pumped into graphite-moderated reactor core from the bottom, where it flows through dozens to hundreds of fuel channels. The fission reaction occurs in the core, within the liquid fuel itself, and then the fission-generated heat is flown out from the top of the core into the heat exchanger. The cooled salt is pumped again into the core, repeating the process. The secondary loop is filled with liquid salt without nuclear fuel, which then transfers the heat from primary loop to the energy conversion system.

As thermal breeder MSR works in thorium fuel cycle, its research typically employs U-233 as its fissile driver [9], [10], [13]–[16], save for studies on the transition to thorium fuel cycle [17]–[20]. However, U-233 is non-existent in nature, and thereby must be synthetised either using a specialised facility like proposed in MSR-FUJI [7] or bred in other reactors [21], [22]. Synthetising U-233 in will typically result in impure U-233. That is, the U-233 is accompanied by other uranium isotopes. Uranium contaminants can alter the neutronic characteristics of thermal breeder MSR, and so far, this issue has not been addressed, including for PCMSR.

The objective of this study is to assess how much U-233 purity affects the neutronic performance of an MSR, where PCMSR was chosen as the simulated MSR model. The assessed parameters are effective multiplication factor (k_{eff}), temperature coefficient of reactivity (TCR), conversion ratio (CR), and kinetic parameters. MCNP6 radiation transport code is used to calculate PCMSR neutronic parameters. This research will be beneficial in deciding whether starting up PCMSR, or any other MSR in general, with impure U-233 makes sense in term of reactor physics performance, and if yes, how much of impurity is acceptable. This will have an impact on the stringency of U-233 synthesis process and its associated production cost.

2. Reactor Description

PCMSR is a molten salt reactor operated in thorium fuel cycle. It employs graphite as moderator and liquid fluoride salt as fuel and coolant. In the core, lithium fluoride (LiF) salt is used as the carrier salt. For the intermediate and tertiary coolant, eutectic Flinak (LiF-NaF-KF) salt is used. Whilst LiF in the core has its lithium-7 isotope enriched to 100%, LiF in secondary salt is unenriched to reduce cost and provide a protective layer to prevent criticality during accident [10].

The original PCMSR was intended to operate at operational temperature of 1373K [10]. In the previous model comparison study, it was reduced to 1200K to adjust with the current nuclear structural material developments [23]. To maintain the same electrical power output, its thermal power was increased to 570 MWt up from 460 MWt. General schematic of PCMSR system is shown in Fig. 1.

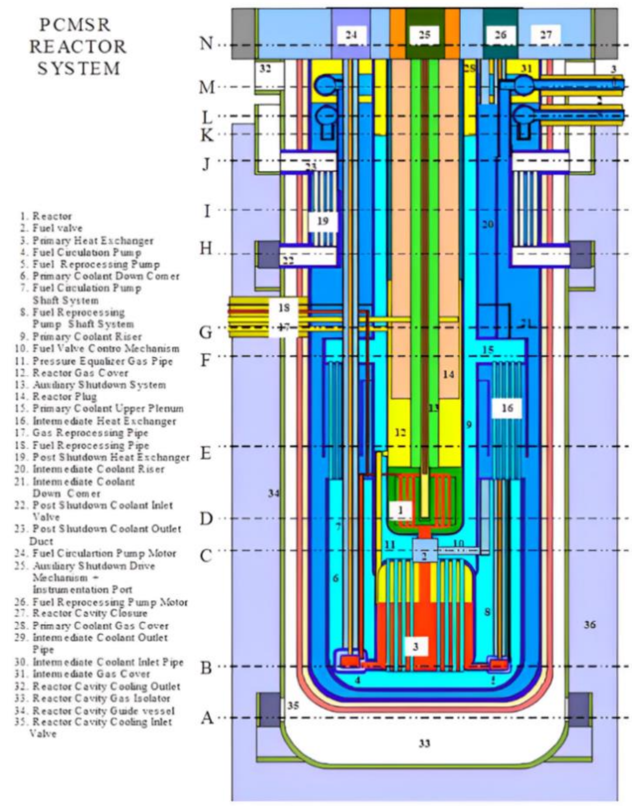

Fig. 1. PCMSR Reactor System [10].

The PCMSR core used in this research was adopted from the aforementioned study [23]. It maintains the configuration as a virtual one-and-half fluid core [3]. It has a single fuel stream, where the fissile and fertile fuel are dissolved in the same fuel mixture. The fuel stream the flows into two separate zones in the reactor core, referred to as "core" and "blanket" zone. The "core" with narrower channel is intended to optimise fission reaction, whilst "blanket" has larger channel diameter to optimise neutron capture by thorium. Two fuel-less graphite layers separate the "core" with control rod layer and blanket zone. The visualisation of PCMSR reactor core in MCNP is displayed in Fig. 2. Meanwhile, the reactor parameters are provided in Table 1.

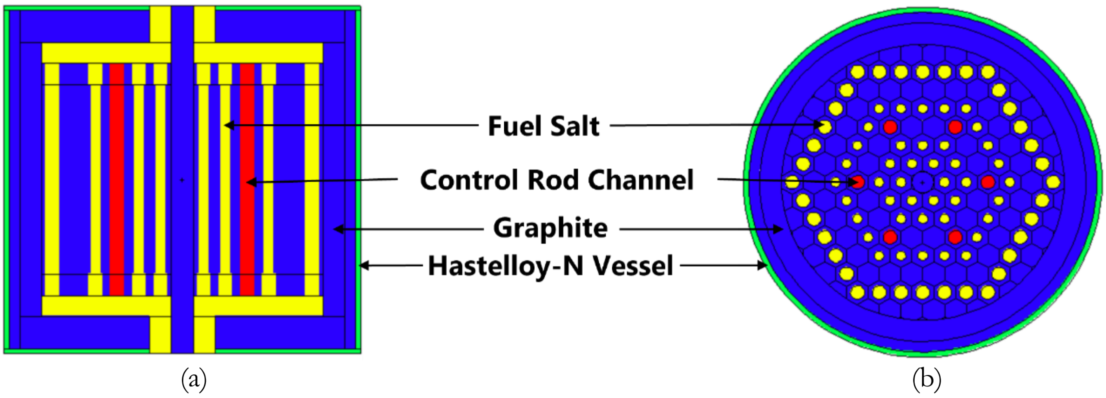

Fig. 2. PCMSR Reactor Core Model (a) Axial view; (b) Radial view.

Table 1. PCMSR reactor parameters [18].

Thermal power	570 MWt
Active core diameter	220 cm
Active core height	220 cm
Graphite density	2.2 g/cm^3
Hastelloy thickness	5 cm
Core channel radius	4.5 cm
Blanket channel radius	6.5 cm
Operational temperature	1200K
Fuel type	Molten salt
Composition	70%LiF-30%(Th-^{233}U)F_4
Fuel density	3.75 g/cm^3

The PCMSR reactor design employed in this study was found to be unable to achieve thermal breeding [23]. Therefore, the reactor is operated as a high converter reactor, with less stringent reprocessing requirement but needs a certain amount of external U-233 supply. Since U-233 is expensive to synthetise, the impact of different fuel purity on the fuel feed requirement is important to be analysed.

3. Methodology

The primary focus of this study was neutronic analysis of PCMSR using U-233 as the fissile driver with different impurities. Here we used four different U-233 vectors, one pure and three impure [21], [22], [24], [25]. Each vector has decreasing level of U-233 purity; Vector 1 is the purest U-233 (100%) whilst Vector 4 has the lowest purity. Table 2 summarised the vector composition of the simulated U-233 fuels.

Table 2. U-233 impurity vectors.

Isotope	Vector 1	Vector 2	Vector 3	Vector 4
U-232	0%	0.00%	0.00%	0.05%
U-233	100%	92.80%	85.20%	63.03%
U-234	0%	6.50%	12.50%	25.12%
U-235	0%	0.65%	2.00%	5.92%
U-236	0%	0.04%	0.20%	5.88%

The scope of neutronic analysis encompassed k_{eff}, TCR, CR, and kinetic parameters. These calculations were performed using Monte Carlo N-Particle version 6 (MCNP6) radiation transport code with ENDF/B-VII.0 continuous neutron cross section library. This radiation transport code is capable to model various physics phenomena involving transport of radiation particle, including reactor physics, medical application, and radiation protection. MCNP has been used and validated for many nuclear reactor analyses [26]–[30]. MCNP6 was previously used for the model comparison of PCMSR [23], also used to model various thermal MSR designs [28], [30]–[35] as well as fast MSR [36]. The research is employed at the beginning of cycle, without considering burnup calculation, since MCNP lacks the defining feature to analyse online fuel reprocessing in MSR.

In calculating core criticality, the aforementioned study maintained the k_{eff} value below $1+\beta_{eff}$. This approach was taken since MSR does not necessarily require high excess reactivity as it can be refuelled online. Since this study compared various fuel compositions, to make it more uniform, the k_{eff} was kept at around 1.005 ± 0.001. Thus, the fuel compositions were adjusted in order to obtain k_{eff} around the said value.

Calculation of kinetic parameters were done using KOPTS card in MCNP6 [37]. The card can generate several kinetic parameters from criticality calculation. Here, the evaluated kinetic parameters were effective delayed neutron fraction (β_{eff}) and average neutron generation time (Λ), which are important parameters in reactor control.

TCR calculation was divided into three components: Doppler coefficient (DC), salt density coefficient (SDC), and moderator temperature coefficient (MTC). DC was calculated by lowering the temperature to 900K without adjusting the fuel density. To calculate SDC, salt density was adjusted but the temperature remained constant. MTC was obtained by lowering the moderator temperature to 900K and adjusting the s(α,β) thermal scattering library to 1000K [38].

CR was calculated using the formula expressed in Eq. 1, modified from [16].

$$CR = \frac{R_C({}^{232}_{90}Th + {}^{232}_{92}U + {}^{234}_{92}U + {}^{238}_{92}U)}{R_A({}^{233}_{92}U + {}^{235}_{92}U)} \quad (1)$$

where R_C is the neutron capture reaction rate of fertile nuclides and R_A is the neutron absorption reaction rate of fissile nuclides. As the CR was calculated from reaction rate, the equation can be applied in the beginning of cycle (BOC) only, since it ignores online fuel reprocessing.

For criticality and TCR calculations, the simulated neutron at each cycle was set at 50,000 neutrons, 250 total cycles with 50 first cycles discarded. This results in standard deviation value of ± 19 pcm for k_{eff} calculations and ± 23 pcm for β_{eff} calculations.

4. Results and Discussion

4.1. Initial Criticality

The result of criticality calculation is provided in Table 3.

Table 3. k_{eff} values for various fuel purity.

	k_{eff}	Fissile %mol
Vector 1	1.00554 ± 0.00019	0.499
Vector 2	1.00512 ± 0.00019	0.512
Vector 3	1.00529 ± 0.00019	0.523
Vector 4	1.00488 ± 0.00019	0.569

Fuel impurity clearly affects the required fissile content in order to achieve the desired k_{eff}. Fissile requirement increases as U-233 purity decreases, primarily due to neutron capture by U-234. The latter transmutes into U-235, whose neutronic performance is less efficient than U-233 due to higher neutron capture cross section, thereby increasing capture-to-fission ratio (see Fig. 3). Fissile requirement is the highest in Vectors 4, being the lowest in U-233 content and highest in U-234 and U-235 contents.

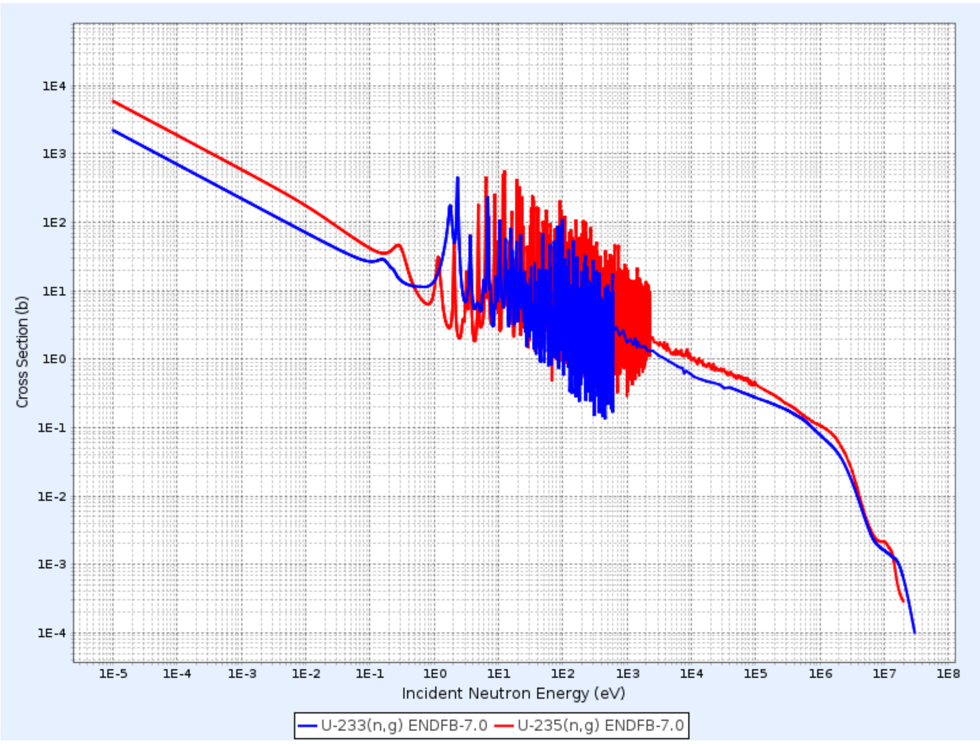

Fig. 3. Neutron capture cross section of U-233 (blue) and U-235 (red). The cross section was taken from Evaluated Nuclear Data Library (ENDF)/B-VII.0.

Compared to Vector 1, impure vectors require 2.59-14.1% more initial U-233, translated into 4.29-23.36 kg U-233. Whilst it does not seem to be particularly large, as previously mentioned, U-233 is expensive to synthetise. Assuming U-233 synthesis cost at USD 240/g [39], an additional cost at more than USD 5 million is required for starting up the reactor (see Table 4 for summary). Even higher initial fuel cost can be incurred if the cost of U-233 synthesis is higher than the aforementioned estimate.

Table 4. Additional Fissile and Cost Relative to Vector 1.

Vector	Additional Fissile (kg)	Additional Initial Fuel Cost (million USD)
2	4.29	1.03
3	4.92	1.96
4	23.36	5.61

4.2. Neutron Spectra

Neutron spectra of PCMSR fuelled with various level of U-233 purity are illustrated in Fig. 4. Various fuel compositions do not appear to give significant spectral difference. Nevertheless, Vector 1 has its thermal flux the highest compared to the rest, whilst Vector 4 is the lowest. This implies that higher U-233 purity resulted in slightly softer neutron spectrum, as the heavier nuclides with high neutron-absorbing capability cause the neutron flux to be suppressed. Nevertheless, the difference is insignificant. It can be concluded that in a fixed core, different U-233 purity does not meaningfully impact neutron spectrum profile.

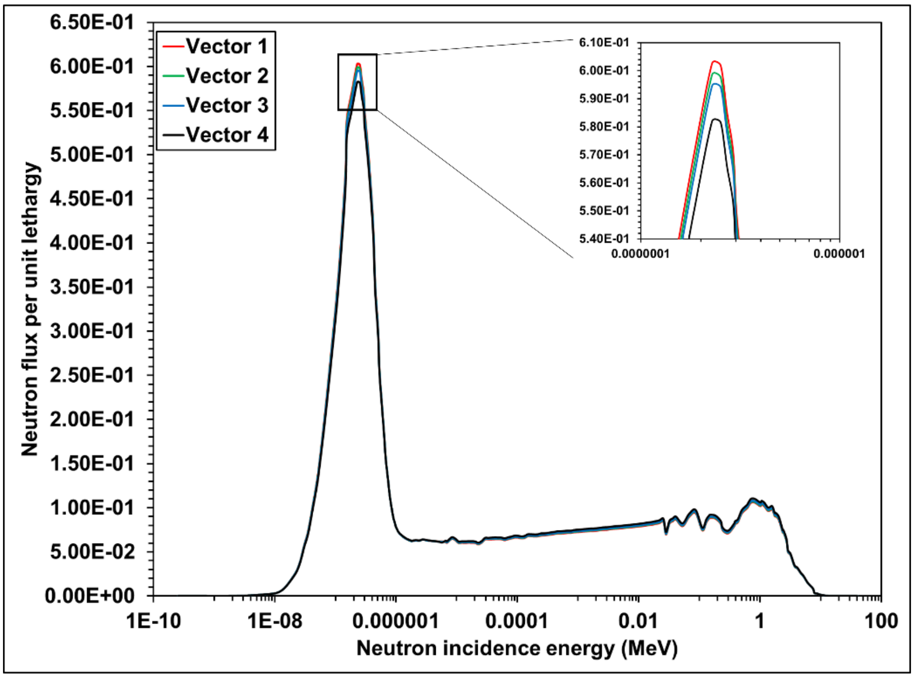

Fig. 4. Neutron flux per unit lethargy of PCMSR with various fuel vectors.

4.3. Kinetic Parameters

Reactor kinetic parameters are displayed in Fig. 5. For β_{eff}, it appears that there is no particular pattern regarding its value against vector composition. Theoretically, with increasing U-235 content, which has significantly higher delayed neutron fraction than U-233, the β_{eff} should be higher as well. However, even in Vector 4, there is no notable difference of β_{eff} value compared to other vectors with lower to no U-235 content. This means that U-235 plays little part in overall fission reaction, which is quite understandable since the highest U-235 content is only slightly lower than 6 mole%.

Considering the standard deviation (± 23 pcm), β_{eff} values remain on the similar region for all vectors. From here, it can be deduced that different U-233 purity has little to no impact to the β_{eff} value. Especially when U-235 content is low, as delayed neutron fraction is highly dependent on fissile nuclide used as the fuel. Vector 4, as seen in Table 2, has low U-235 content despite being highly impure, for U-234 is the second most dominant isotope. The latter has little usefulness to the neutronic of U-233 fuelled reactors, since it has negligible thermal fission cross section [40]. However, it has a decent neutron capture cross section.

On the other hand, Λ value is decreasing with the increase of U-233 impurity. As impure vectors require higher fissile content in order to achieve criticality, the atomic density of fissile nuclides in the reactor core are also increasing. This shortens the time for fission neutrons to roam the reactor core, as the probability of the neutrons being absorbed by fissile nuclides is higher. Nonetheless, the maximum difference in Λ is around 5.84% for Vector 4 against Vector 1, which has the longest Λ.

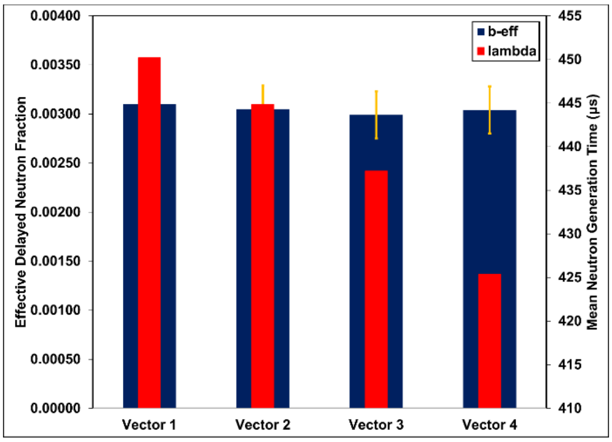

Fig. 5. Kinetic parameters of PCMSR with various fuel vectors.

In term of reactor control, shorter Λ would be more difficult to control, especially if the TCR is not particularly negative. Fortunately, Λ values of PCMSR are comparably longer than LWR [41] and generally similar with HTGR [37]. This is due to graphite moderator has lower neutron moderating power than light water, so that more collisions with graphite atoms are needed to thermalise the fission neutron. MSR can also control reactivity by adjusting fuel flow debit into the core, owing to its liquid fuel characteristics. Generally, kinetic parameters are not significantly altered by U-233 impurity.

4.4. Temperature Coefficient of Reactivity

TCR is the most important inherent safety characteristics in MSR. Result of TCR calculation is provided in Fig. 6.

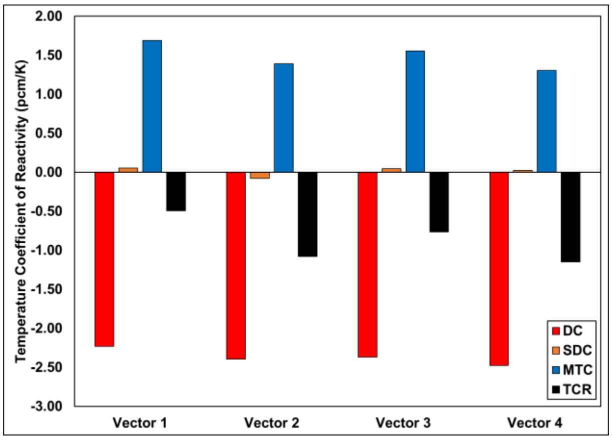

Fig. 6. Temperature coefficient of reactivity for PCMSR with different U-233 purity.

Unlike kinetic parameters, TCR values show a peculiar characteristic. The pattern in DC is irregular, although typically its negative value strengthened with the increase of fissile content in the core. This occurs due to the obtained k_{eff} value is not necessarily identical, the largest reactivity difference being 66 pcm between the highest (Vector 1) and lowest (Vector 4). With the reactivity difference between Vector 1 and Vector 3 is lower than Vector 1 and Vector 2, the DC is more negative in Vector 2 than Vector 3 (see Table 5). From this finding, in a comparative analysis using identical geometry, the change in DC value is highly sensitive to the difference of k_{eff} value. The larger the difference, the more impactful the DC change of value to overall TCR value.

Table 5. Reactivity and Doppler coefficient difference for various U-233 purity.

Vector	Difference in reactivity value, relative to Vector 1 (pcm)	Difference in DC value, relative to Vector 1
2	-42	-0.161
3	-25	-0.134
4	-66	-0.244

DC values of PCMSR are significantly less negative than other MSRs [15], [16], [42], [43]. Such phenomenon is likely caused by the small core size of PCMSR. It hardens the neutron spectrum, and thus reducing the effect of Doppler broadening. Small fuel salt volume also contributed, as the fuel loading within the core is also smaller, in spite of higher fuel fraction.

SDC shows an unusual pattern in Vector 2. Whilst the other vectors, including Vector 1, have slightly positive SDC, which implies that the reactor core leans more onto under-moderated region, Vector 2 shows a slightly negative SDC. This might result from calculation uncertainty, as Vector 3 and 4 show no regular pattern either. Overall, since all SDC values are close to zero, the reactor core geometry is more or less at optimum moderator-to-fuel ratio (MFR), the latter implies a balanced core moderation, neither overly moderated nor less moderated than necessary.

Meanwhile, MTC values are all positive. This is a common phenomenon in MSRs fuelled by U-233 caused by spectral shift of the graphite moderator when it is heated. Although PCMSR uses high-density graphite, the MTC values for all vectors are not too dissimilar to other thermal breeder MSRs in various studies [15], [16], [42], [43]. U-235 content increases, positive value of MTC is slightly weakened, due to graphite spectral shift does not affect U-235 fission rate. The pattern, again, is irregular, due to the same reason with DC values.

In total, due to some irregularities, the total TCR value shows no apparent pattern except the TCR value improved with the increase of U-233 impurity. A maximum of 57% TCR improvement was found in Vector 4, contributed from less positive MTC and more negative DC. Compared to other parameters already discussed, U-233 purity has more impact on this inherent safety parameter. The primary disadvantage of MSR fuelled with U-233 is the weak TCR contributed by the positive MTC. Although MSR core with low U-233 purity contains higher U-233 content, the co-existence of other impurities and fissile nuclide (U-235) readily compensate the positive moderator feedback against temperature. This shows that U-233 impurities are actually beneficial in improving the safety aspect of MSR, especially since the TCR typically degrades throughout operation due to the change of nuclides composition inside the fuel.

4.5. Conversion Ratio

CR denotes the ability of a reactor to convert fertile nuclides into fissile nuclides over fissile consumption through fission and capture. Previous study [23] shows that original PCMSR cannot achieve breeding due to ineffective moderation and high neutron leakage. In theory, U-233 impurity will cause the CR to be reduced even further. However, the analysis shows a different light on this matter. CR calculation for four U-233 vectors can be seen in Fig. 7.

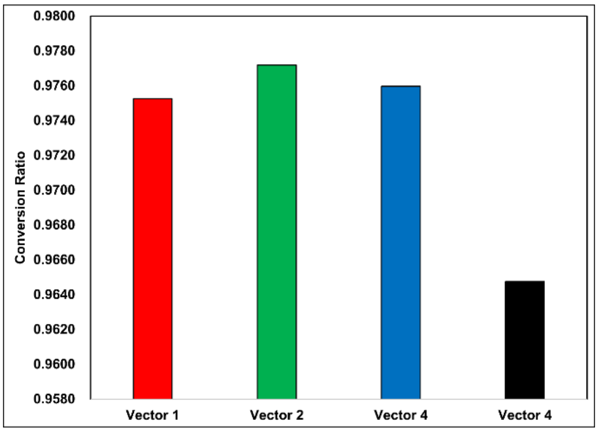

Fig. 7. Conversion ratio of PCMSR with various fissile vector.

The interesting finding from Fig. 7 is that Vector 1 has lower CR than Vectors 2 and 3, albeit the difference is tremendously small. Such result is caused by the existence of U-234 in impure vectors, itself being a decent neutron absorber, as previously mentioned. Additional U-234 capture increased fertile conversion and somehow compensated the lower thorium capture due to larger fissile fraction. This behaviour was also observed in author's previous work [34]. In that work, the CR value of MSR fuelled with reactor grade plutonium, which is less pure than weapon grade plutonium, was higher. In that case, abundant fertile fuel Pu-240 in reactor grade plutonium acts as the strong neutron absorber that results in an increased CR.

Nevertheless, in the long term, thorium capture plays more crucial role in maintaining reactor criticality, since U-233 has lower capture-to-fission ratio than U-235 [44]. This means that U-233 has higher probability to induce fission reaction and releases energy instead of simply capturing neutrons and transmutes into a fertile uranium isotope. Therefore, even if Vector 1 has slightly lower CR, the theory is that it will be compensated after a certain period of burnup. This study omitted burnup calculation, and thus the evolution of fissile materials cannot be completely verified. Since PCMSR is not a breeder reactor, it needs a constant supply of external fissile to maintain criticality, albeit the requirement is extremely small due to its high CR [45].

Meanwhile, Vector 4 is found to be having the lowest CR. Large U-234 fraction in the fuel is proved to be unable to compensate the lower thorium capture, and subsequently the CR value is decreased. This implies that, even if there is a slight improvement of CR in Vectors 2 and 3, there should be a point where the improvement peaked as neutron economy worsen with the increase of U-234 content and the decrease of thorium content. Although the absolute difference is insignificant, the difference in breeding gain (1 – CR) is more prominent. As seen from Table 6, breeding gain in Vector 4 is significantly lower than Vector 1, resulting in a higher U-233 fissile feed requirement. Due to high CR, however, the additional fissile requirement is marginal. Notwithstanding, more analysis regarding nuclides evolution throughout burnup must be investigated in future works to provide better insight.

Table 6. Breeding gain and additional fissile difference for various fissile purity.

Vector	Difference in breeding gain, relative to Vector 1	Additional fissile difference, relative to Vector 1 (kg)
2	8%	-0.82
3	3%	-0.30
4	-42%	4.45

In breeder MSR, the breeding ratio (BR) evolves throughout burnup process, initially increasing from the initial BR value, and then decreasing afterwards until an equilibrium BR is achieved [9], [43], [46]. Those calculations were performed assuming a 100% pure U-233 was used at the start-up. Over time, the purity of U-233 in the core will degrade as other uranium isotopes build up [18]. If a low-purity U-233 is used as the start-up fuel, apart from the decrease of initial BR value, the U-233 purity will be degraded even further and reduces neutronic performance throughout its operational time. The consequence is the longer doubling time.

Further studies can be directed to observe the impact of U-233 purity to the MSR performance more comprehensively, by performing burnup calculation and using a thermal breeder MSR model as a complementary of high converter model. This will enhance the understanding of reactor core behaviour when the MSR is fuelled with impure U-233. Therefore, a proper reactor fuelling strategy, which considers technological and economic feasibility of U-233 synthesis, can be developed and implemented for each type of MSR.

5. Conclusions

The comparative assessment on PCMSR neutronic performance using various U-233 purity vectors resulted in several findings. First, U-233 impurity insignificantly affects kinetic parameters and has marginal impact on CR. Although impure vectors have lower Λ, the difference is relatively small. Second, higher fissile fraction requirement in impure U-233 will increase the initial fissile inventory, but its effect on plant economics should not be detrimental. Third, U-233 impurity has higher impact on TCR, generally improving the inherent safety feature by weakening the positive MTC and strengthening the negative DC. Overall, using impure U-233 as the initial fissile fuel for PCMSR may actually be beneficial in improving its safety aspect. Nonetheless, extremely low U-233 purity is detrimental to the fissile breeding performance, and thereby the minimum U-233 purity should be set at a medium (± 85% U-233). By using medium U-233 purity, the cost of external fuel feed can be reduced as the U-233 synthesis process can be made less stringent and the TCR can be improved ever since the beginning of operation, ensuring a safer reactor response to reactivity-initiated accidents.

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Author Contribution Statement

R. Andika Putra Dwijayanto: Conceptualisation, Methodology, Formal Analysis, Writing – Original Draft. Andang Widi Harto: Software, Supervision, Writing – Review and Editing. R. Andika Putra Dwijayanto is the main contributor of the manuscript. All authors read and approve the final version of the paper.

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