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Investigation on Inherent Safety of One Fluid-Molten Salt Reactor (OF-MSR) with Various Starting Fuel

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Molten salt reactor (MSR) is often associated with thorium fuel cycle, thanks to its excellent neutron economy and online reprocessing capability. However, since ^{233}U, the fissile used in pure thorium fuel cycle, is not commercially available, the MSR must be started with other fissile nuclides. Different fissile yields different inherent safety characteristics, and thus must be assessed accordingly. This paper investigates the inherent safety aspects of one fluid MSR (OF-MSR) using various fissile fuel, namely low-enriched uranium (LEU), reactor grade plutonium (RGPu), and reactor grade plutonium + minor actinides (PuMA). The calculation was performed using MCNPX2.6.0 programme with ENDF/B-VII library. Parameters assessed are temperature coefficient of reactivity (TCR) and void coefficient of reactivity (VCR). The result shows that TCR for LEU, RGPu, and PuMA are -3.13 pcm, -2.02 pcm and -1.79 pcm, respectively. Meanwhile, the VCR is negative only for LEU, whilst RGPu and PuMA suffer from positive void reactivity. Therefore, for the OF-MSR design used in this study, LEU is the only safe option as OF-MSR starting fuel.

1. INTRODUCTION

Molten salt reactor (MSR) has recently gained popularity as a prospective Generation IV nuclear reactor technology. MSR differs from conventional light water reactor (LWR) in term of usage of graphite moderator and liquid fuel in form of salt compound instead of solid oxide fuel. Fluoride salt has high boiling point, ensuring that MSR can operate in high temperature (700°C and above) without pressure vessel. This feature enhances plant safety and reducing cost [1–3]. MSR is also capable of online fuel reprocessing, thanks to its liquid fuel. This feature is ideal to utilise thorium, which technically capable of breeding in thermal spectrum. Thus, many MSR concepts are designed to use thorium in its fuel [4–7].

MSR operating in pure thorium fuel cycle requires thorium as its fertile fuel and ^{233}U as its fissile fuel [4, 6]. The latter is not naturally occurring, since its half-life (160,000 years) is comparably shorter than the only naturally occurring fissile nuclide, ^{235}U (703.8 million years). However, MSR can be started by virtually any fissile isotope whilst gradually transition into pure thorium cycle [8]. This includes ^{235}U and ^{239}Pu. The latter is often considered as a potential proliferation threat, despite plutonium isotopes in LWR spent fuel is degraded so much that it is unsuitable for military diversion. Burning plutonium in MSR can help eliminating the perceived threat [9].

The issue is that different fissile nuclide used can result in different neutronic behaviour, including the inherent safety [10, 11]. This is important since, in term of fuel utilisation, both ^{235}U and ^{239}Pu are inferior than that of ^{233}U in thermal spectrum. It is also suggested that, to

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prevent proliferation potential even further, that plutonium isotopes are not to be separated from its accompanying minor actinides (MA) [9, 12]. Thus, it is also possible that the plutonium used as startup fuel of MSR contains MA.

This study investigated the inherent safety parameters of One Fluid-Molten Salt Reactor (OF-MSR) using various starting fissile fuels, namely low-enriched uranium (LEU), reactor grade plutonium (RGPu), and reactor grade plutonium + minor actinides (PuMA). The investigation is important to understand the behaviour of aforementioned fissile options in an MSR, since different fissile fuel can lead into different neutronic safety characteristics. Inherent safety parameters were discussed in many previous studies with different reactor designs [10, 11, 13, 14]. This study would employ a One Fluid-Molten Salt Reactor (OF-MSR), adapted from earlier design of Passive Compact Molten Salt Reactor (PCMSR) [15]. The calculation was performed using MCNPX2.6.0 transport code with ENDF/B-VII continuous neutron library.

2. GENERAL DESCRIPTION

OF-MSR has its fertile and fissile nuclides dissolved within a single carrier salt. Such configuration is simpler than its two-fluid counterpart, at the expense of lower inherent safety [16]. Eutectic lithium fluoride-beryllium fluoride salt, colloquially known as Flibe salt, is employed in this study, as it is considered as the best carrier salt for MSR. Thorium, along with fissile LEU, RGPu, and PuMA are dissolved in Flibe salt and circulated in the primary system.

The general design of the OF-MSR is adopted from early design of PCMSR, with several design modifications. Although the fluid stream is singular, it has two moderation zones like MSBR. The narrower fuel channel is intended as 'core,' where fissile reaction is dominant. Meanwhile, the wider fuel channels act as 'blanket' zone, where thorium capture is more dominant than in the 'core.' This configuration is confirmed in a study [13] as more effective in fuel breeding compared to pure one fluid MSR.

OF-MSR core parameters are provided in Table 1. Core configuration is kept the same with PCMSR. The core is divided into three zones; upper and lower zone are 'blanket' area, whilst middle zone is the 'core' area. Modifications are made at larger core dimension, significantly lower operational temperature (630 °C as opposed to 1200°C), core and blanket channel radius, and added radial boron carbide layer for protection against radiation damage to the Hastelloy vessel which shrouded the core.

The axial cross section of OF-MSR core generated in MCNPX is shown in Fig. 1.

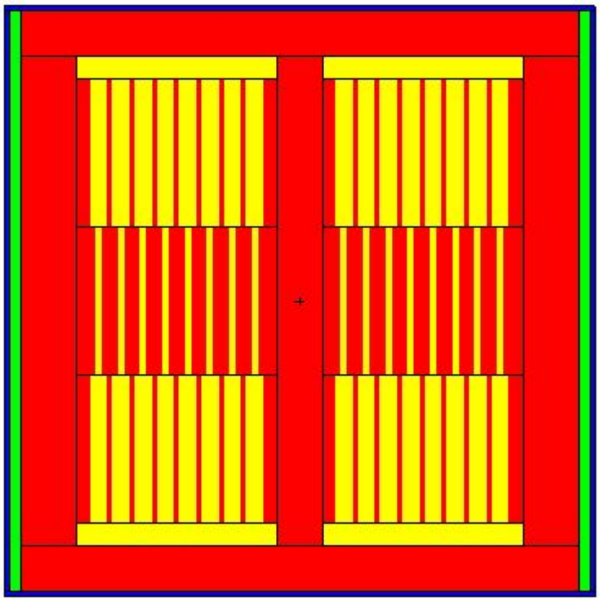

Fig. 1. MCNPX model on axial cross section of OF-MSR

Table 1. OF-MSR core parameters [9, 15, 17]

Active core diameter	400 cm
Active core height	400 cm
Graphite density	1.84 g/cm^3
Hastelloy thickness	10 cm
Core channel radius	3 cm
Blanket channel radius	8 cm
Operational temperature	630 °C
Fuel type	Molten salt
Composition	LiF-BeF_2-ThF_4-$^{238}UF_4$-$^{235}UF_4$ LiF-BeF_2-ThF_4-PuF_3 LiF-BeF_2-ThF_4-PuF_3-MA
Plutonium isotopic vector	^{238}Pu-^{239}Pu-^{240}Pu-^{241}Pu-^{242}Pu
Plutonium vector composition (%)	1,58-57,76-26,57-8,76-5,33
Minor actinide vector	^{237}Np-^{241}Am-^{243}Am-^{242}Cm-^{243}Cm-^{244}Cm-^{245}Cm-^{246}Cm
Minor actinide vector composition (%)	42,25-47,57-8,5-0,32-0,01-1,26-0,07-0,01

The fuel is thorium and fissile nuclides dissolved within a Flibe salt. The composition is set at 70% LiF, 15% BeF_2, and 15% $(HM)F_x$. The heavy metal (HM) consists of fertile ThF_4 and fissile either UF_4, PuF_3, or PuF_3+$(MA)F_3$. LEU enrichment is set at 20%, to minimise ^{238}U content in the fuel. Plutonium and MA vectors are taken from reference [9]. As for PuF_3+$(MA)F_3$, the Pu:MA ratio is set at 9:1.

3. METHODOLOGY

Calculation of inherent safety was performed using MCNPX2.6.0 neutron transport code with ENDF/B-VII continuous neutron energy library. KCODE module was employed to calculate criticality at steady-state condition. Although MSR fuel is circulating instead of static, previous study has validated the use of MCNP to model MSR [18]. Neutrons simulated at each cycle was set at 10,000 neutrons, with total of 220 cycles and the first 20 cycles were skipped.

OF-MSR does not require high excess reactivity due to its online refuelling capability. Excess reactivity can then be maintained below delayed neutron fraction (β) of each fissile nuclide [11]. Since MSR fuel is circulating, a portion of β is "lost" from the core, reducing its value. MCNPX2.6.0 cannot calculate the lost β, and thus the original β is used anyway.

The first calculation was performed to determine the critical mass of OF-MSR system. The k_{eff} value is maintained below 1+β. After the critical composition was obtained, the second calculation was performed to determine temperature coefficient of reactivity (TCR). This calculation is important since OF-MSR often suffers from small negative TCR and even positive, an undesirable trait.

Calculation by Oak Ridge National Laboratory (ORNL) showed that two-fluid MSR has more negative temperature coefficient of reactivity (TCR) [16] compared to OF-MSR, in this case the Molten Salt Breeder Reactor (MSBR) [19]. Another study even doubted whether OF-MSR has negative TCR at all [20]. More recent study indicates that OF-MSR can achieve negative TCR, despite not as negative as LWR or two-fluid MSR [21]. This undesirable trait particularly analysed for pure thorium cycle. Nevertheless, it would be worthwhile to investigate whether this trait exists when using other fissile fuel

Temperature simulated in this paper are at 600K, 900K, and 1200K, with 900K is set at the operating temperature. Liquid fuel expands when heated, reducing its density and pushing fuel out of the core. Thus, TCR in OF-MSR is not only a factor of Doppler broadening, but also liquid fuel expansion. Fuel density is corrected for each temperature. This paper addressed TCR as a total core TCR.

In MSR, fuel density change can be treated as a void, apart from gaseous fission product buildup in the core or offgas bubbling. Thus, void coefficient of reactivity (VCR) of OF-MSR is calculated by reducing the density of fuel salt. Density reduction is set from 0-50%.

4. RESULTS AND DISCUSSION

Criticality calculation was performed until the OF-MSR core met critical condition, with the excess reactivity is kept below 1+β for each fissile nuclide. OF-MSR typically requires 0.2-0.4% mole of fissile fuel in order to be critical, depending on core geometry and fuel composition used [22]. Tables 2 and 3 show the criticality value of each fissile nuclide and its molar composition. Molar percentage are below 0.4% for all fissile nuclides. Thus, the model can be considered as valid.

Table 2. k_{eff} value for various fissile fuel at 900K

Fuel	Molar composition (%)	k_{eff}
$LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$	70-15-13.14-1.86	1.00494 ± 0.00042
$LiF\text{-}BeF_2\text{-}ThF_4\text{-}PuF_3$	70-15-14.6-0.43	1.00071 ± 0.00047
$LiF\text{-}BeF_2\text{-}ThF_4\text{-}PuF_3$-MA	70-15-14.4-0.574-0.6374	1.00126 ± 0.00051

Table 3. Molar composition of OF-MSR fissile fuel

Fuel	Isotopic Vector	Molar Composition
LEU	$^{238}U\text{-}^{235}U$	1.49-0.37
RGPu	$^{238}Pu\text{-}^{239}Pu\text{-}^{240}Pu\text{-}^{241}Pu\text{-}^{242}Pu$	0.01-0.25-0.11-0.04-0.02
PuMA	$^{238}Pu\text{-}^{239}Pu\text{-}^{240}Pu\text{-}^{241}Pu\text{-}^{242}Pu\text{-}$	0.01-0.33-0.15-0.05-0.03-
	$^{237}Np\text{-}^{241}Am\text{-}^{243}Am\text{-}^{242}Cm\text{-}$	0.02693-0.03033-0.00542-0.0002
	$^{243}Cm\text{-}^{244}Cm\text{-}^{245}Cm\text{-}^{246}Cm$	0.00001-0.0008-0.00004-0.00001

Theoretically, ^{239}Pu is worse in thermal spectrum compared to ^{235}U. This is due to the higher capture-to-fission ratio of ^{239}Pu. Thus, it was initially expected that fissile ^{239}Pu required in order to be critical is larger than that of ^{235}U. However, Table 3 shows otherwise. Whilst ^{235}U required to be critical is 0.37% mole, only 0.25% mol (0.29% mol with ^{241}Pu) is required for RGPu to make the reactor critical. This is by taking into account that RGPu only uses thorium as fertile, as opposed to LEU which inevitably contains ^{238}U. The latter has smaller capture cross section, so that theoretically also smaller fissile concentration is required to make a ^{238}U-containing system critical. Such phenomenon does not appear in this case.

Similar phenomenon occurred in PuMA. Although total fissile nuclide required to make the OF-MSR critical is slightly higher than LEU (0.38% mol), MA is a strong neutron absorber, most notably ^{237}Np and ^{241}Am which exist in considerable amount in the fuel. Consequently, high concentration of plutonium is needed. Yet, the required fissile is only differs slightly with that of LEU. These issues with Pu-based fuel may have something to do with the geometry, as will be discussed later on.

After critical composition is determined, the following step is to determine the TCR value. To comply with safety criteria, the reactor must be designed is such a way so that the TCR is negative. With negative TCR, the reactor will self-regulate when power fluctuations occur, maintaining its stability. Although it was mentioned previously that OF-MSR somehow suffers from less negative TCR, it mainly occurs for pure thorium cycle, and may not applied to other fuel composition.

TCR for various fuel composition are shown in Figs. 2, 3, and 4.

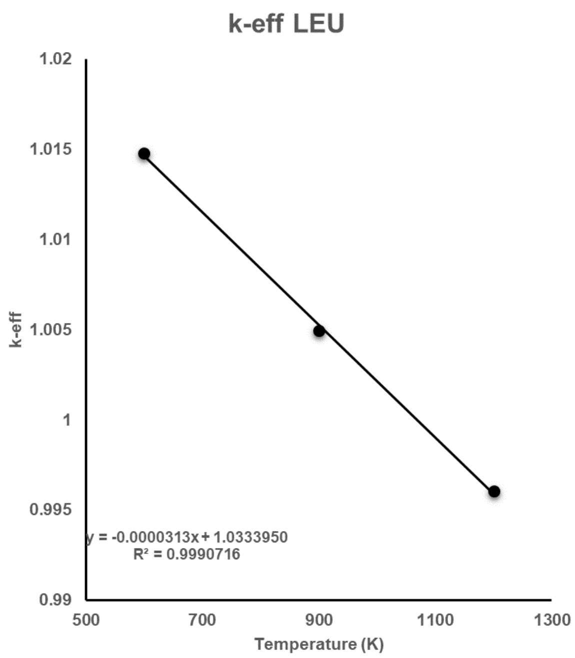

Fig. 2. k_{eff} change against temperature for LEU

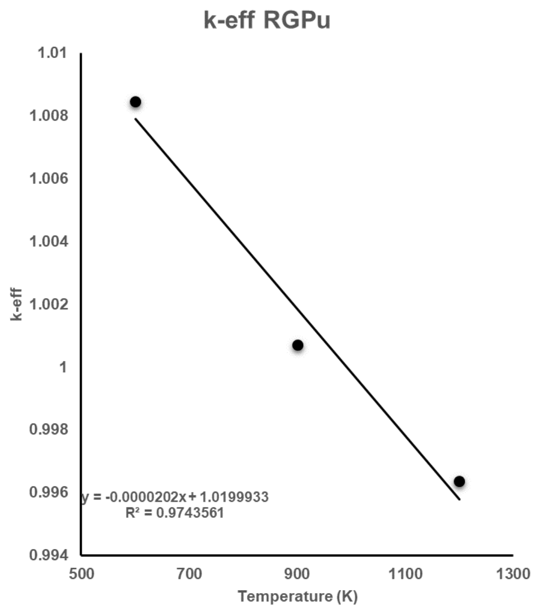

Fig. 3. k_{eff} change against temperature for RGPu

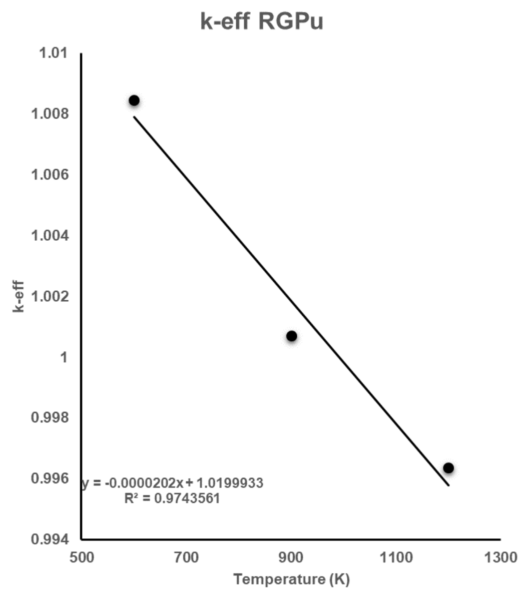

Fig. 4. k_{eff} change against temperature for PuMA

TCR values are negative for all starting fuel options. Therefore, the issue with pure thorium cycle does not appear when the OF-MSR is started with fissile fuel other than ^{233}U. Aside from Doppler effect, negative TCR is also induced by thermal expansion of molten salt. LEU has the most negative TCR, around -3.13 pcm/K. Although graphite moderator is known to be providing positive reactivity feedback, Doppler effect combined with salt expansion is sufficient to provide total negative reactivity. Coexistence of ^{232}Th and ^{238}U may also play a part, since ^{238}U introduction adds more resonance peaks, consequently reducing resonance escape probability and in turn enhances the Doppler effect.

TCR value of RGPu and PuMA are -2.02 pcm/K and -1.79 pcm/K, respectively. Apart from the harder neutron spectrum, the absence of ^{238}U can explain the less negative TCR of plutonium-fuelled OF-MSR compared to LEU. Meanwhile, inclusion of MA in the fuel salt proved to be providing positive temperature coefficient. When the salt expands, strong neutron absorbers such as ^{237}Np and ^{241}Am are partially ejected from the core, thus lowering neutron capture. This in turn increases core reactivity.

Another possible reason to explain the less negative TCR found in RGPu and PuMA is the moderating condition. As opposed to conventional light water reactor (LWR), MSR is operated in over moderated condition. This is due to the phase of fuel and moderator is reversed; liquid fuel with solid moderator. In under moderated condition, when the salt expands, moderator-to-fuel ratio increases. It resulted in softened neutron spectrum. This is further exacerbated by positive reactivity feedback from graphite. Thus, in under moderated condition, the fuel reactivity feedback can be positive instead of negative.

Moderating condition of OF-MSR is a factor of core salt volume, in turn determined by the core channel radius. Larger core channel radius reduces moderator-to-fuel ratio, hardening the spectrum and increasing fissile content. Plutonium has lower tolerance for higher core salt volume, since its spectrum is already hard compared to uranium anyway. Meanwhile, uranium isotopes such as ^{233}U and ^{235}U can maintain being in over moderated condition with larger core channel radius. In this case, core channel radius of 3 cm is possibly in the under moderated region for RGPu and PuMA.

Despite total TCR is still negative, provided mainly by Doppler broadening, this is by no means that RGPu and PuMA are automatically safe in term of VCR. In MSR, void is usually formed by fission products in gaseous form. Helium sparging to remove noble gas fission products such as xenon and krypton can also cause void.

K_{eff} value for each fuel is shown in Figs. 5, 6, and 7.

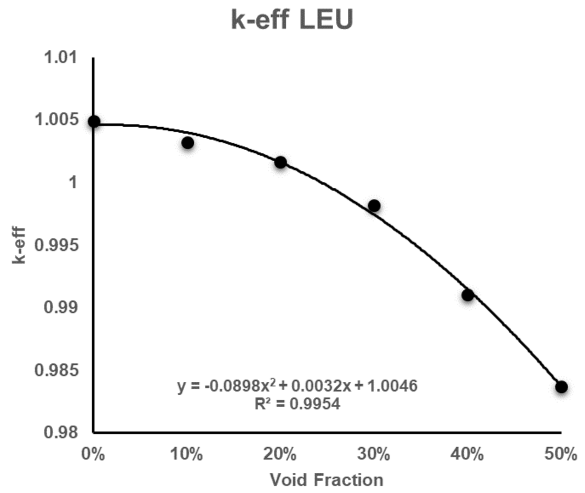

Fig. 5. LEU criticality against void fraction at 900K

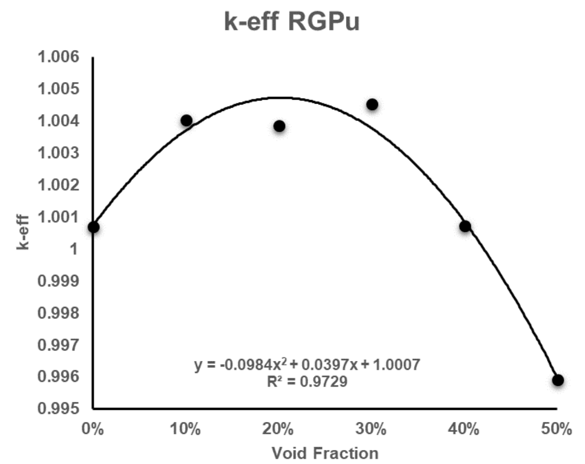

Fig. 6. RGPu criticality against void fraction at 900K

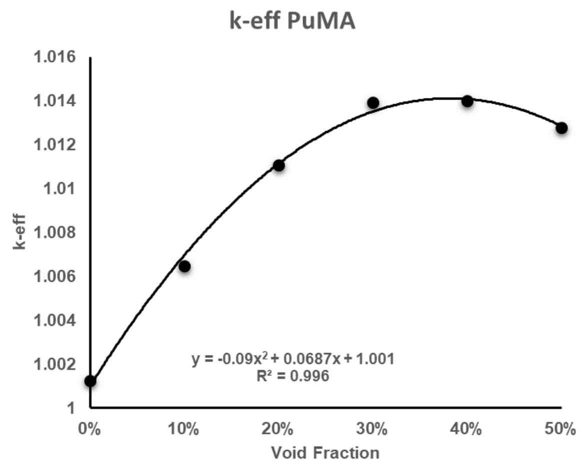

Fig. 7. PuMA criticality against void fraction at 900K

Among the options, only LEU proves to be inherently safe. At every void fraction, its k_{eff} decreasing. It implies that LEU-fuelled is in over moderated zone. Thus, LEU satisfies negative VCR criteria. However, the same condition cannot be necessarily attributed to RGPu and PuMA.

In RGPu, void caused the k_{eff} to increase up until 30% fraction, before finally decreased and subcritical in 50% fraction. Thus, for the same core channel radius, RGPu is already shifted into under moderated condition. Even worse is shown in PuMA; although it similarly peaked in 30% fraction, the k_{eff} did not decrease significantly at 50% fraction. Moreover, k_{eff} increase in PuMA is more than twice as large as RGPu. Meaning, addition of MA in the studied core geometry shifts the moderation condition quite extremely. This is understandable, since PuMA has the hardest spectrum among the fuel options, as shown in Fig. 8.

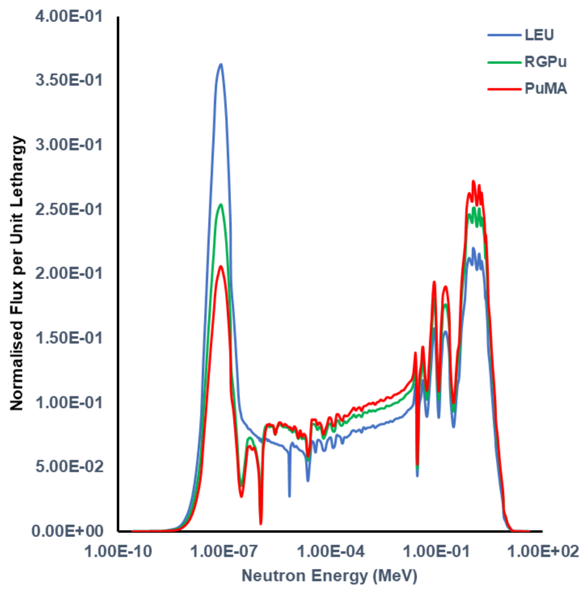

Fig. 8. Neutron spectrum of the various OF-MSR startup fuel

Neutron spectrum of LEU-fuelled OF-MSR is softer compared to RGPu, and even more so compared to PuMA. At under moderated condition, when the fuel salt expands, neutron spectrum in RGPu and PuMA tend to soften as moderator-to-fuel ratio increases, which favour fission over capture. The effect is even more apparent in PuMA, as mentioned previously, since neutron-absorbing MA such as ^{237}Np and ^{241}Am are partially ejected from the core.

Hardened neutron spectrum can also explains the less fissile requirement for RGPu and PuMA. Both ^{239}Pu and ^{241}Pu emit more neutrons in fast spectrum compared to ^{235}U. Their fast capture cross section is also smaller than fission cross section, whilst ^{235}U is the reverse. Since RGPu and PuMA both have higher peak in fast spectrum, they increase neutron population and fast fission, as an addition to the thermal fission. Thus, core critical mass is reduced. In a glance, this might contradict the fact that plutonium do not perform particularly well in thermal spectrum. However, it is also a proof that the core is in under moderated condition, which is not the ideal condition for MSR operation. Geometry dependence of core critical mass and its correlation with moderating condition must be investigated in the future works.

K_{eff} change against void is not linear. Thus, linear progression is unsuitable to determine average VCR value. VCR is instead calculated at each step of void fraction change. The results are provided in Table 4.

Table 4. VCR value of each fissile composition (pcm/%void)

Void fraction change	LEU	RGPu	PuMA
0-10%	-17.36	33.04	51.80
10-20%	-15.33	-1.79	45.20
20-30%	-34.50	6.64	27.99
30-40%	-72.58	-37.80	0.39
40-50%	-74.88	-48.16	-11.78

5. CONCLUSION

In a fixed geometry, OF-MSR shows different characteristics of inherent safety when using different starting fuels. LEU as starting fuel can achieve negative TCR and VCR, thereby satisfies inherent safety criteria and suitable as starting fuel. On the other hand, although RGPu and PuMA both achieve negative TCR, albeit less negative than that of LEU, the VCR values are positive until 30% void fraction. Thus, RGPu and PuMA are inherently unsafe. However, these safety characteristics are limited to the geometry used in this study and cannot be necessarily generalised to other OF-MSR designs. As inherent safety characteristic is a factor of moderator-to-fuel ratio, in order to ensure that the reactor is inherently safe, OF-MSR must be designed with appropriate core channel radius, i.e. the optimum design. This notion must be assessed in the future works.

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