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NUCLIDE COMPOSITION ANALYSIS OF PCMSR FUEL USING THORIUM AS SUSTAINABLE FUEL AND LOW ENRICH URANIUM AS STARTING FUEL

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ABSTRACT

Passive Compact Molten Salt Reactor (PCMSR) is an advanced nuclear reactor designed to utilize thorium as the main fertile fuel to achieve sustainable long term natural nuclear fuel resources utilization. Thorium (i.e. ^{232}Th) will be converted to ^{233}U and finally ^{233}U undergoes neutron induced fission reaction and generates reactor thermal power. Because U-233 does not exist at initial fuel, the other fissile fuel is needed for initial operation of PCMSR. To avoid illicit use of high grade fissile materials, the low enrich uranium (LEU) is used for the initial fissile fuel. During several years of the initial reactor operation, ^{235}U at LEU will be depleted and ^{233}U will be produced and also be consumed. To maintain reactor criticality, the production of ^{233}U must balance the consumption of ^{233}U and the depletion of ^{235}U. For long term operation, the reactor must be able to maintain its criticality by solely the balance of ^{233}U production and ^{233}U consumption. This paper explains the numerical study results of the ability of the PCMSR to maintain its fuel composition. The PCMSR in this study uses the initial fuel with the composition of 6.3 % mole (LEU)F_4, 23.7 % mole of $^{232}ThF_4$ and 70 mole % of ^{7}LiF. The enrichment level of LEU is 19 % mole of ^{235}U. The nuclide composition of the PCMSR fuel will be analyzed by solving the simultaneous ordinary differential equations of nuclide balances. There are 38 nuclides involved in this analysis including uranium isotopes, plutonium isotopes, minor actinides (Np, Am, Cm), thorium, protactinium and six neutron poison fission product nuclides. The calculations are performed for several values of specific power i.e.: 9 MWth/(ton HM), 18 MWth/(ton HM), 27 MWth/(ton HM), 36 MWth/(ton HM) and 44 MWth/(ton HM). The fuel extraction rate is assumed to be proportional to the specific power values, i.e 931 (cm^3/day)/(MWth/(ton HM)). The specific thorium injection rate is 828 kg/GWey. The calculation results show that the PCMSR fuel can achieve quasi equilibrium fuel composition for long term operation (i.e. 100 years). In this condition, the total fissile mole fraction (i.e. the sum of mole fractions of ^{233}U and ^{235}U related to total heavy metal mole) can be maintained to almost constant value (i.e. at the range of 2.600 to 2.759). However during the early years of its operation, there is a depression of the value of total fissile mole fraction. The depression becomes more prominent as the value of specific power increases. The minimum fissile mole fraction is 2.477 for the specific power of 9 MWth/(ton HM), 2.430 for 18 MWth/(ton HM), 2.365 for 27 MWth/(ton HM), 2.304 for 36 MWth/(ton HM), 2.237 for 44 MWth/(ton HM). This depression is due to the delay time of the production of ^{233}U after the neutron capture of ^{232}Th. Based on these calculations, a low specific power is recommended. The calculation results can be summarized that for long term operation, PCMSR fuel can maintain total fissile mole fraction at quasi equilibrium condition. A depression of total fissile mole fraction value occurs during the early years of PCMSR fuel operation, which becomes more prominent as the increasing value specific power.

Keywords: PCMSR fuel, thorium sustainable fuel, LEU starting fuel, quasi equilibrium composition.

INTRODUCTION

Nuclear energy technology will become prominent future energy technology to replace conventional (fossil fuel) energy technology. The nuclear energy technology has capability to produce output energy in massive and continuous manner. The massive and continuous energy supply is one of important factors to increase national industrial level.

The main problems of the recent nuclear energy technology are fuel resources sustainability, very long term high level radioactive and possibility of nuclear fuel proliferation.

Recent nuclear reactors are mostly non breeder reactors and using ^{235}U as the effective nuclear fuel. The mole fraction of ^{235}U in natural uranium resources is only 0.71. It means that the recent nuclear reactors use only 0.6 % - 0.7 % of the overall nuclear fuel resources.

The amount of the known proven natural uranium recently is 5000 kilo tons [1]. The recent worldwide nuclear reactor capacity power is roughly 390 GWe [2]. Based on these, the worldwide known proven nuclear fuel resources is estimated to be available only for the next 50 until 80 years [3].

The fact that the recent nuclear reactor technology uses only 0.6 % - 0.7 % of the overall nuclear fuel resources means that more than 99 % of the nuclear material is useless or become waste.

Due to the enrichment process, 88 % of the natural uranium resources becomes unusable depleted uranium (DU) and only 12 % becomes useful low enrichment uranium (LEU). The recent nuclear reactor technology can only utilize 5 % of nuclear fuel material in LEU. A fuel stream balance calculation [4] shows that the recent nuclear reactor technology produces 20 – 30 tonne per GWe year of nuclear waste in the form of spent fuel.

The next generation of nuclear reactor must be designed to use the rest of natural uranium content (^{238}U), to reuse spent fuel of recent reactor; or to use the more abundant nuclear fuel resource, *i.e.* thorium [5]. To achieve these capability, the next generation nuclear reactors must have breeding capability. With breeding capability, all nuclear material resources can be used in such reactor. The consequence, the nuclear fuel utility factor can be increased with the factor of 150 times. It means that the recently known nuclear fuel resources will be available for more than 1000 years [4].

FUNDAMENTAL THEORY AND METHODOLOGY

a) PCMSR (Passive compact molten salt reactor)

PCMSR (Passive Compact Molten Salt Reactor) is designed to use thorium (^{232}Th) as sustainable fuel [6]. In operation, ^{232}Th will be converted to ^{233}U by the reactor itself. The exact fuel composition depends on the initial fuel composition (i.e. depends on the fuel fabrication) and the on line fuel reprocessing during operation [7]. PCMSR use graphite as moderator and structure and euthectic flinak salt (LiF-NaK-KF) as intermediate coolant [6].

The use of molten salt fuel combined with graphite moderator gives some important benefits, i.e.: low pressure and high temperature operation [8], high breeding ratio [9], inherent safe [9], high thermal efficiency [8], flexibility in output energy utilization [9] and produce less minor actinides than the use of ^{238}U as fertile fuel [10].

To start PCMSR operation, initial fissile fuel is needed. Lucotte *et al.* have performed a study of using ^{233}U and a mixture of Pu and minor actinide recovered from spent fuel of the recent reactor spent fuel as initial fuel for TMSR (Thorium Molten Salt Reactor) [10]. The ^{233}U, however, does not exist naturally. The use of ^{233}U can be performed only if a previous thorium breeder reactor has existed, thus it cannot be performed for the first thorium breeder reactor. Moreover, ^{233}U and recovered Pu are among high grade fissile materials that have capability to be used as nuclear weapon [11]. Avoiding the use of high grade fissile material will increase proliferation resistant.

In this study, low enrich uranium (LEU) with enrichment level below 20 % mole of ^{235}U will be chosen as the initial fissile fuel of PCMSR. The ability of the reactor to maintain nuclide composition in the fuel is the aims of this study.

Figure-1 shows the overall diagram of PCMSR system including the reactor module and the turbine module. The turbine system produces output power of 250 MWe and the reactor thermal power is 460 MWth (thermodynamic efficiency = 55 %).

PCMSR reactor module is an integral system consists of reactor, fuel circulation system, primary heat exchanger, passive primary coolant circulation system, intermediate heat exchanger, post shutdown heat exchanger and passive post shutdown intermediate coolant circulation system All of the reactor module components are arranged inside a reactor system integral vessel as shown in Figure-2.

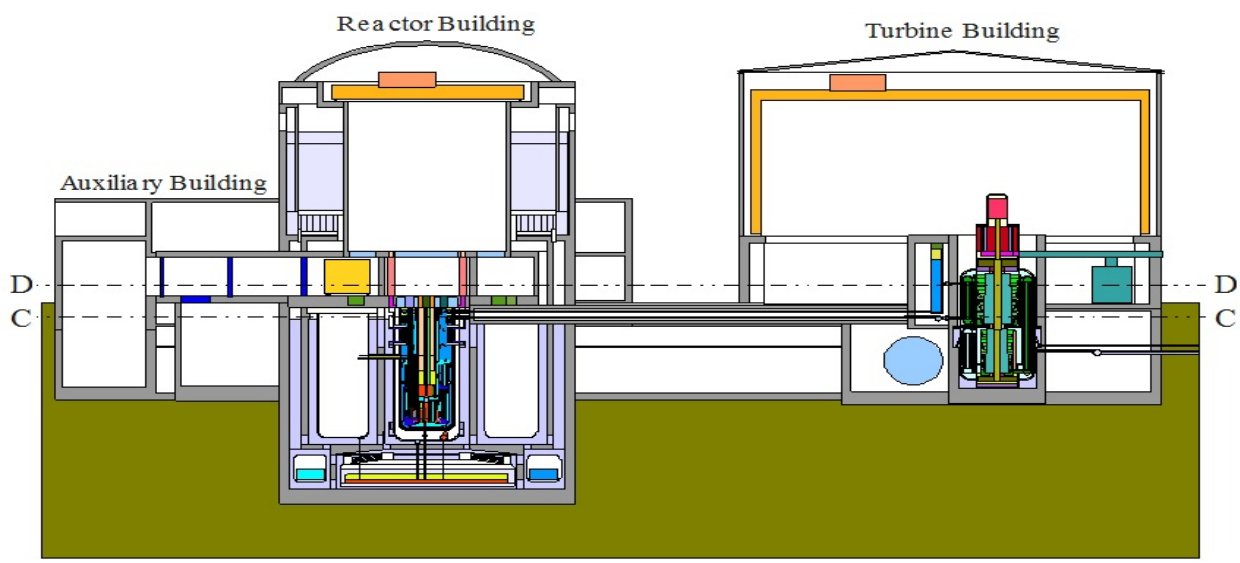

Figure-1. Overall configuration of modular 250 MWe PCMSR system [6].

b) The PCMSR on line fuel processing system

On line fuel processing system is the key for successful operation of PCMSR with sustainable thorium based fuel cycle. Figure-3 shows the schematic diagram of the PCMSR on line refueling and on line fuel processing system. The fuel processing system will extract Pa in the first extractor and fission products in the second extractor. The fission product is then sent to a waste processing

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system. The 233Pa is sent to Pa decay tank to decay to become 233U. The 233U is then removed from the Pa decay tank and re-injected back to the reactor core.

Thorium is continually added to compensate 232Th that converted to 233U during reactor operation. Thus the mole fraction of 233Th can be maintained almost constant value. The LEU is however not added. Both of 235U and 238U will deplete. The 235U will deplete faster than 238U. For sustainable operation, the depletion of 235U must be balanced by the build up of 233U. Meanwhile, the rest of 238U is utilized to prevent the mole fraction of total fissile U (233U and 235U) to LEU level (below 20 %).

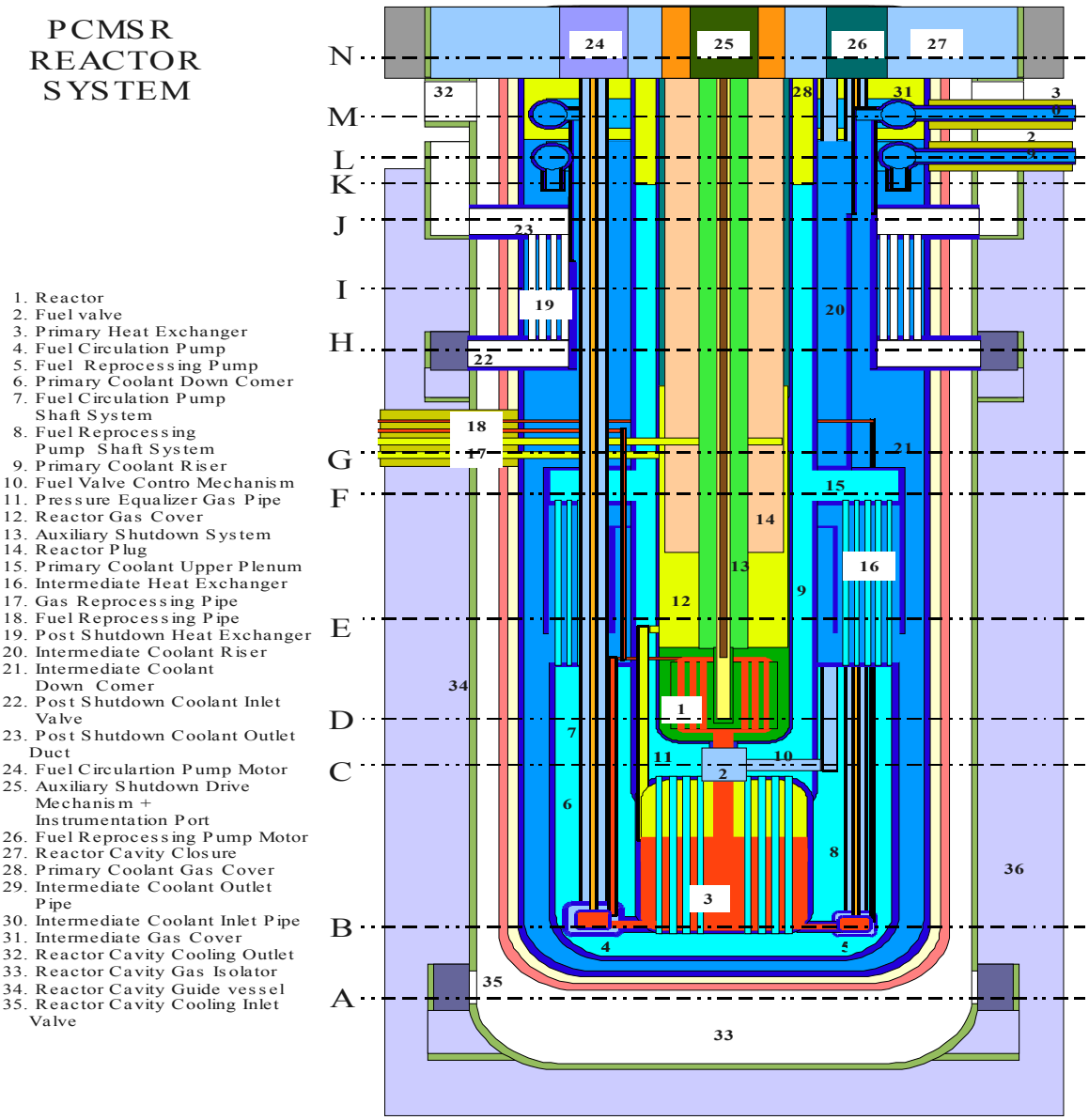

Figure-2. PCMSR reactor module [6].

c) Method

Fuel composition of PCMSR will change dynamically with time until equilibrium condition is reached. The fact that the fuel continuously flows out from and flows in into the reactor core must be considered fuel composition calculation. The nuclide transmutation chain of PCMSR fuel is shown at Figure-4.

Due to the continue mixing, it is assumed that the fuel composition throughout the reactor core is uniform. By this assumption, the equation of the dynamics fuel composition can be expressed as follow:

$$\frac{d}{dt}\left(V_C\Omega_{FC}+V_{B1}\Omega_{FB1}+V_{B2}\Omega_{FB2}+V_H\Omega_{FH}+V_E\right)N_{Z,A}$$
$$=\sum_{i=1}^{I}Q_{in,i}N_{in,i,Z,A}-\sum_{k}^{K}Q_{out,k}N_{Z,A}$$
$$-N_{Z,A}\sum_{g=1}^{G}\sigma_{a,g,Z,A}\left(\int_{V_C}\phi_{gC}\zeta_{C,g}dV_C+\int_{V_{B1}}\phi_{gB1}\zeta_{B1,g}dV_{B1}+\int_{V_{B2}}\phi_{gB2}\zeta_{B2,g}dV_{B2}\right)+$$
$$\begin{pmatrix}-\lambda_{Z,A}N_{Z,A}+\lambda_{Z-1,A}N_{Z-1,A}f_{\beta^-,Z,A-1}+\lambda_{Z+1,A}N_{Z+1,A}f_{\beta^+,Z+1,A}\\+\lambda_{Z+2,A+4}N_{Z+2,A+4}f_{\alpha,Z+2,A+4}\\+\lambda_{Z,A+1}N_{Z,A+1}f_{n,Z,A+1}+\lambda_{Z,A+2}N_{Z,A+2}f_{2n,Z,A+2}\end{pmatrix}\begin{pmatrix}V_C\Omega_{FC}+V_{B1}\Omega_{FB1}+\\V_{B2}\Omega_{FB2}+V_H\Omega_{FH}+V_E\end{pmatrix} \quad (1)$$
$$+N_{Z,A-1}\sum_{g=1}^{G}\left(\sigma_{a,g,Z,A-1}-\sigma_{f,g,Z,A-1}\right)\left(\int_{V_C}\phi_{gC}\zeta_{C,g}dV_C+\int_{V_{B1}}\phi_{gB1}\zeta_{B1,g}dV_{B1}+\int_{V_{B2}}\phi_{gB2}\zeta_{B2,g}dV_{B2}\right)$$
$$+\sum_{j=1}^{J}N_{Z_j,A_j}\sum_{g=1}^{G}y_{Z,A,Z_j,A_j,g}\sigma_{f,g,Z_j,A_j}\left(\int_{V_C}\phi_{gC}\zeta_{C,g}dV_C+\int_{V_{B1}}\phi_{gB1}\zeta_{B1,g}dV_{B1}+\int_{V_{B2}}\phi_{gB2}\zeta_{B2,g}dV_{B2}\right)$$

where :

A	Mass Number	Z	Atomic number
g	Index of group energy of neutron	G	Number of neutron energy group
i	Index of inlet flow to reactor system	I	Number of inlet flow to reactor system
k	Index of outlet flow from reactor system	K	Number of outlet flow to reactor system
j	Index if fissionable nuclei	J	Number of fissionable nuclei
V_C	Reactor core volume (cm^3)	V_{B1}	First blanket volume (cm^3)
V_{B2}	Second blanket volume (cm^3)	V_H	Primary heat exchanger volume (cm^3)
Ω_{FC}	Fuel volume fraction of reactor core	Ω_{FB1}	Fuel volume fraction of first blanket
Ω_{FB2}	Fuel volume fraction of second blanket	Ω_{FH}	Fuel volume fraction of primary heat exchanger
N	Nuclide density (nuclide/cm^3)	f_{β^-}	Negative beta decay fraction
σ_a	Microscopic neutron absorption cross section (cm^2)	f_{β^+}	Positive beta decay fraction
σ_f	Microscopic fission cross section (cm^2)	f_α	Alpha decay fraction
y	Fission product yield	f_n	One neutron emission decay fraction
λ	Decay constant (s^{-1})	f_{2n}	Two neutrons emission decay fraction
ζ_C	Neutron flux depression factor of core	ϕ_{gC}	Average neutron flux in reactor core (n/(cm^2.s))
ζ_{B1}	Flux depression factor of first blanket	ϕ_{gB1}	Average neutron flux in first blanket (n/(cm^2.s))
ζ_{B2}	Flux depression factor of second blanket	ϕ_{gB2}	Average neutron flux in second blanket (n/(cm^2.s))
Q_{in}	Fuel inlet volumetric flow rate (cm^3/s)	Q_{out}	Fuel outlet volumetric flow rate (cm^3/s)

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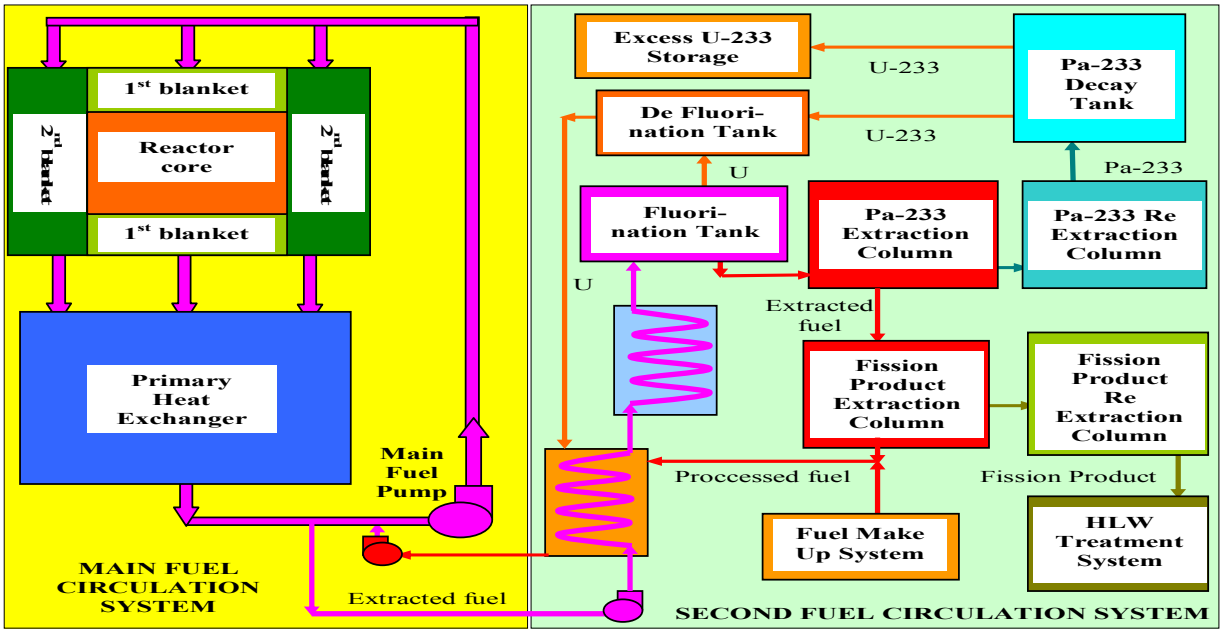

Figure-3. A schematic diagram of the PCMSR on line refueling and on line fuel reprocessing system [6] (with modification).

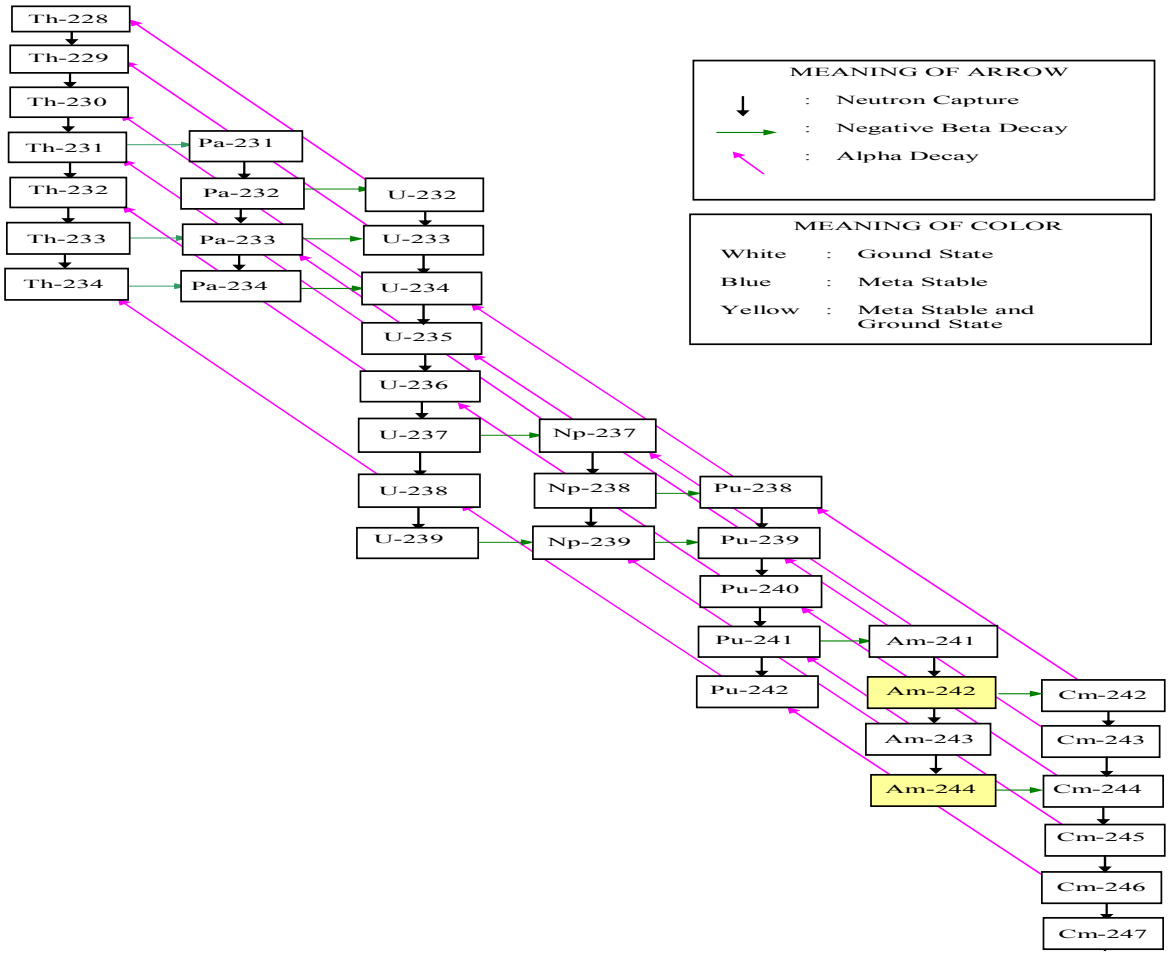

Figure-4. Nuclide transmutation chain of the PCMSR fuel.

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The equation (1) is formulated for all nuclides involved in PCMSR reactor fuel. There are 38 nuclides involved in this analysis including uranium isotopes, plutonium isotopes, minor actinides (Np, Am, Cm), thorium, protactinium and six neutron poison fission product nuclides. The nuclear cross section and decay data is adopted from JANIS 2.1 integral nuclear data software. The efficiency of both of the Pa-extraction system and fission product extraction system is assumed to be 95 %.

The nuclide composition of the PCMSR fuel will be analyzed by solving the simultaneous of these ordinary differential equations of nuclide balances. The interval integration method is used to solve the equations. The calculations are performed for several values of specific power i.e.: 9 MWth/(ton HM), 18 MWth/(ton HM), 27 MWth/(ton HM), 36 MWth/(ton HM) and 44 MWth/(ton HM). The fuel extraction rate is assumed to be proportional to the specific power values, i.e 931 (cm3/day)/(MWth/(ton HM)). The specific thorium injection rate is 828 kg/GWey.

RESULTS AND DISCUSSION

a) Fissile mole fraction

Figure-5 shows the mole fraction of 235U for 100 years of PCMSR fuel operation for several values of the reactor specific power.

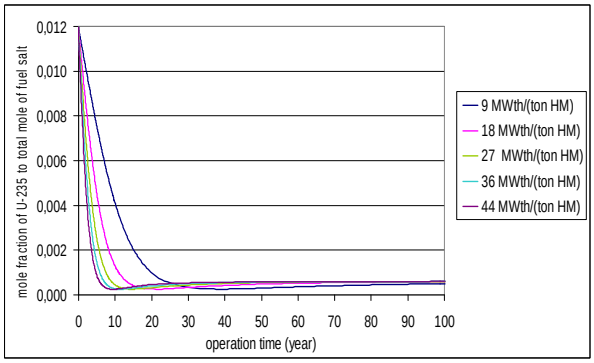

Figure 5. Mole fraction of 235U to total mole of fuel salt in PCMSR fuel [6].

The 235U initially exist as initial fuel loading as part of initial LEU. There is no additional of LEU, thus both of 235U and 238U are depleted as the reactor operation time increase. The 235U will be depleted faster as the reactor specific power increases. The higher specific value means higher neutron flux. The higher neutron flux means higher fission and neutron capture reaction rate by the 235U. As a consequence, 235U is consumed rapidly at higher reactor specific power.

The build up of 233U is shown at Figure-6 for several values of the reactor specific power. The 233U is produced after 232Th absorbs neutron. The 232Th is then transmuted to 233Th, which it quickly decays to 233Pa. The 233Pa then decays more slowly to become 233U.

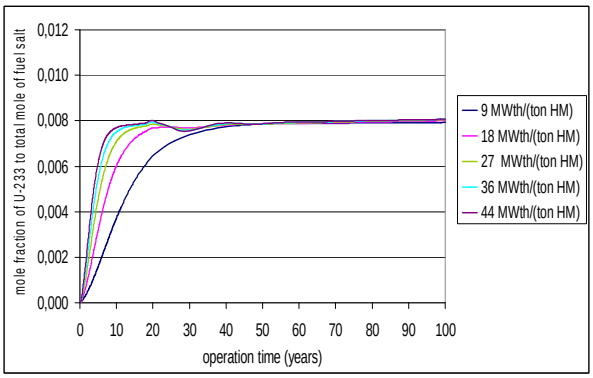

Figure-6. Mole fraction of 233U in to total mole of fuel salt PCMSR fuel [6].

Figure-6 shows that at early years of PCMSR operation time, the mole fraction of 233U increases. The increasing of 233U is more rapid as the specific reactor power increases. This is due to the fact that increasing the specific reactor power will increase neutron flux and in turn will increase the production of 233U by neutron capture of 232Th.

The increasing of mole fraction of 233U will stop as the production of 233U by neutron capture of 232Th is balanced by the consumption of 233U by fission and neutron capture reaction.

Figure-7 shows the mole fraction of total fissile nuclides (i.e. dominated by the sum of 233U mole fraction and 235U mole fraction). As shown in Figure 5, the amount of 235U at long time operation is very small, thus the fissile material is dominated only by 233U. The PCMSR can maintain almost constant value of total mole fraction of fissile nuclides.

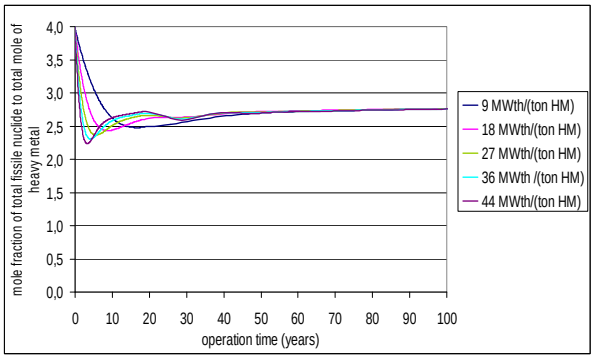

Figure-7. Mole fraction of total fissile nuclide per total mole of heavy metal in fuel salt.

Figure-7 shows that, in this condition, the total fissile mole fraction (i.e. the sum of mole fractions of 233U and 235U related to total heavy metal mole) can be maintained to almost constant value (i.e. at the range of 2.600 to 2.759).

During early time of operation, the total mole fraction of fissile nuclides depends on the existence of 233U and 235U. The two steps of beta decay of 233Th

until become 233U gives a delay time in producing 233U. The delay time makes the rate of 233U is slightly slower than the rate of 235U consumption.

The consequence is the depression of the total mole fraction of fissile nuclide during early time of reactor operation. The depression becomes more prominent as the value of specific power increases. This is due to the more rapid depletion of 235U as the reactor specific time increase.

The minimum fissile mole fraction is 2.477 for the specific power of 9 MWth/(ton HM), 2.430 for 18 MWth/(ton HM), 2.365 for 27 MWth/(ton HM), 2.304 for 36 MWth/(ton HM), 2.237 for 44 MWth/(ton HM). Based on these calculations, the low specific power is recommended to avoid large reactivity swing and to ensure that the reactor can achieve critical condition during all of its operational period.

b) The role of the rest of 238U

The 238U is introduced in reactor as part of LEU at initial reactor fuel. There is no 238U anymore after that during all time of reactor operation. The 238U will deplete as it absorbs neutrons. Because 238U has lower neutron absorption cross section than 235U, and also 238U exist in larger amount than 235U, the 238U depletes far more slowly than 235U. Figure-8 shows the mole fraction of 238U in PCMSR fuel.

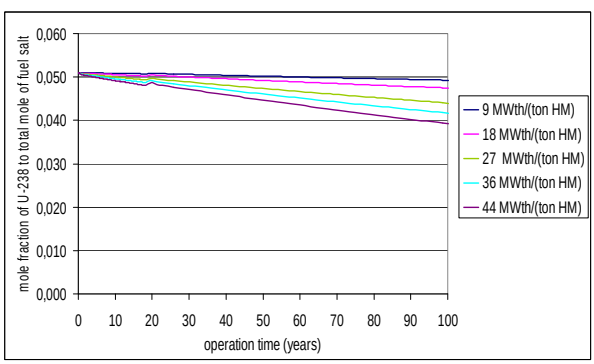

Figure 8. Mole fraction of ^{238}U in to total mole of fuel salt PCMSR fuel.

Figure-8 shows that depletion of ^{238}U is faster as the reactor specific value increases. The reason is similar to the reason for ^{235}U depletion, i.e. the higher the specific power, the higher the neutron flux and thus the higher the ^{238}U reaction rate.

The rest of ^{238}U has important role in NPT point of view. The rest of ^{238}U role is to "hide" or "conceal" the fissile uranium nuclides. This means the existence of ^{238}U has important role to keep the mole fraction of fissile uranium nuclides (i.e. uranium enrichment level) below 20% to fulfill the category of low enrich uranium (LEU). Figure-9 shows the uranium enrichment level, i.e. the molar ratio of fissile uranium (^{233}U and ^{235}U) to total uranium, of PCMSR fuel during its operation time periods.

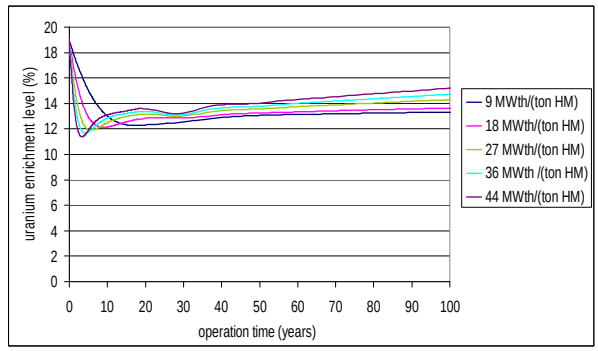

Figure-9. Uranium enrichment level of PCMSR fuel.

Figure-9 shows that the uranium enrichment level decreases rapidly at the early years of the reactor operation. This is due to the depletion of 235U as shown at Figure-5. Figure-9 shows the decrease rate of uranium enrichment level is higher for higher specific power. This is also consistent with the fact that 235U depletes more rapidly as the specific power increases.

As for the quasi equilibrium condition, the amount of 235U becomes negligible and the fissile nuclides are dominated by 233U. The mole fraction of 233U can be maintained at almost constant value due to the balance of 233U and 233U consumption. However, the uranium enrichment level increases due to the depletion of 238U. The increasing of the uranium enrichment level is higher for higher specific power because 238U depletes more rapidly at higher reactor specific power.

The Figure-9 shows that the uranium enrichment level can be kept below 20 % for all time during reactor operation period. It means that the PCMSR design can fulfill NPT requirement by keeping the uranium to stay at LEU condition for all time during reactor operation period.

CONCLUSIONS

This study shows that it is possible for PCMSR reactor using thorium as sustainable fuel and LEU for starting fuel to maintain the amount of fissile nuclides to a certain level (i.e. at the range of 2.600 to 2.759) for long term operation time. However, there is a depression of the amount of fissile nuclide during early time of its operation. The depression becomes more prominent as the reactor specific power increases, thus the low specific power operation is recommended. The PCMSR can fulfill NPT requirement for all operation time because the uranium enrichment level can be kept below 20% to fulfill LEU category.

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