DRAFT (MC2023 Conference Proceedings): Reactor Physics Assessment of Annular Plutonium-Thorium Fuels for Use in Prismatic Fuel Blocks in a HTGR-SMR with a Hydrogen-Based Moderator...

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Reactor Physics Assessment of Annular Plutonium-Thorium Fuels for Use in Prismatic Fuel Blocks in a HTGR-SMR with a Hydrogen-Based Moderator (7LiH)

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ABSTRACT

Small Modular Reactors (SMRs) based on high-temperature gas-cooled reactor (HTGR) technology are of growing interest for potential use in small and remote grids and high-temperature process heat applications. Most HTGR-SMR designs use high-assay low enriched uranium (HALEU) fuel in the form of TRISO fuel particles, and are moderated by graphite. However, there are alternative design concepts for an HTGR-SMR that may offer superior performance characteristics, while utilizing an alternative fissile fuel supply option. In this exploratory study, lattice physics calculations were performed with SERPENT to evaluate an alternative HTGR-SMR prismatic fuel block concept using coated annular fuel pellets instead of TRISO-particle fuel compacts, along with the use of hydrogen-based solid moderator rods made of ^{7}LiH. Different plutonium-thorium oxy-carbide fuels were tested, using either reactor-grade plutonium from recycled pressurized water reactor (PWR) or pressure-tube heavy water reactor (PT-HWR) fuels. Results demonstrate that such fuels can achieve burnups ranging from 35 to 95 MWd/kgIHM, and a core lifetime of ranging from 10 to 35 years, using a 3-batch refueling scheme in a HTGR-SMR. The fissile content in the spent fuel (> 3 wt% fissile/IHM) is high enough that it could be recycled into a larger scale PT-HWR with no need for additional fissile fuel.

KEYWORDS: HTGR, SMR, Plutonium, Thorium, Monte Carlo

1. INTRODUCTION

Small modular reactors (SMRs) based on high temperature gas-cooled reactor (HTGRs) technology are being developed to generate electricity and high-temperature process heat for a variety of applications [1]. Many HTGR-SMR concepts have fissile fuel consumption requirements that are substantially greater than those of pressurized water reactors (PWRs) or pressure tube heavy water reactors (PT-HWRs) per unit energy generated [2]. The greater fuel requirements are due to the use of graphite moderator, which is not as effective as hydrogen-based moderators for small cores, and TRi-structural ISOtropic (TRISO) fuel, which has a lower fissile density than conventional fuel pellets.

A TRISO particle comprises a tiny (~0.02 cm radius) spherical fuel kernel that is surrounded by layers of buffer and pyrolytic carbon and silicon carbide, which are designed to prevent the release of fission products (FPs) at high temperatures (700°C - 1600°C). This design, with multiple layers and coatings of non-fuel structural materials, limits the fissile loading density (# fissile atoms/cm^3), and the mass of fissile fuel that can be loaded in a HTGR core. Hence, uranium enrichments of between 10 and 20 wt% $^{235}U/U$ are usually required to achieve acceptable levels of burnup and core operating life before refueling is required. Alternatives to TRISO fuel in prismatic block HTGRs have been proposed that can reduce fissile consumption dramatically by substituting TRISO particle-embedded compacts with cylindrical fuel pellets that more closely resemble conventional PWR and PT-HWR fuel [3,4]. The results presented in Reference [4] indicate that the use of cylindrical fuel pellets combined with the inclusion of hydrogen-based moderator in the fuel assembly can reduce fissile consumption by up to 72%, and extend the fuel residence time by more than 10 years. This fuel concept is SiC clad, and includes layers of pyrolytic and low-density buffer carbon to retain fission products at high temperatures and burnups, although its performance relative to that of TRISO fuel pellets is a topic for future research.

While HTGRs are primarily being developed to use enriched uranium fuel, previous computational studies have analyzed the performance of TRISO fueled HTGRs as plutonium burners [5-7]. The purpose of this computational study is to evaluate the performance of HTGRs with fuel pellets for burning plutonium with respect to fuel consumption and spent fuel (SF) composition. In this study, the multi-layer annular fuel pellet concept proposed previously in [4] is used, where the enriched uranium is substituted with a mixture of plutonium and thorium.

2. ANNULAR FUEL PELLET CONCEPT

The annular fuel pellet concept that was introduced in [4] is comprised of concentric, cylindrical, layers, a diagram of which is shown in Figure 1. The data in TABLE I shows the material and radii of each layer. The fuel is in the form of an oxy-carbide, such as (Pu,Th)CO, ThCO, or (DU,Th)CO. The volume of oxycarbide fuel in the annular fuel pellet is greater than that of the oxycarbide fuel kernels in TRISO particles in a MHTGR-350 fuel compact [8] by a factor of 14.9. The prismatic fuel block in which the annular fuel is inserted is shown in Figure 2, which is based on the MHTGR-350 fuel block [8]. The modified fuel block includes hydrogen-based moderator rods to compensate for the increased volume of fuel, which provide sufficient additional moderation to achieve high fissile utilization. Specifically, the moderator rods are composed of lithium hydride (^{7}LiH), with the lithium enriched to 99.995 at% $^{7}Li/Li$. Note that the average temperature of the MHTGR-350 fuel block is below 875 K [4], which is less than the melting point of LiH (961 K). It is preferable that LiH remain solid during operation because it begins to decompose when it melts, which would require hydrogen pressurization to prevent its release.

Four configurations of this concept are evaluated, which are listed in TABLE II. Three of these configurations, 10Pu+Th, 20Pu+Th|Th, and 20Pu+Th|DU+Th, have an equivalent Pu content of 10 wt% Pu/IHM, and which differ amongst themselves with respect to the amount of Pu in the inner and outer annuli as well as the presence of depleted uranium (DU). DU (0.2 wt% U-235/U) is blended with Th to denature the U-233 that is produced. The other configuration has 20 wt% Pu/IHM.

Furthermore, two types of plutonium are considered: reactor-grade plutonium from a PWR, which is 67.2 wt% Pu-fissile/Pu, and reactor-grade plutonium from a PT-HWR, which is 72.5 wt% Pu-fissile/Pu. Accumulating reserves of spent fuel from PWRs and PT-HWRs are considered a potentially important source of fissile fuel for supporting a fleet of HTGR-SMRs.

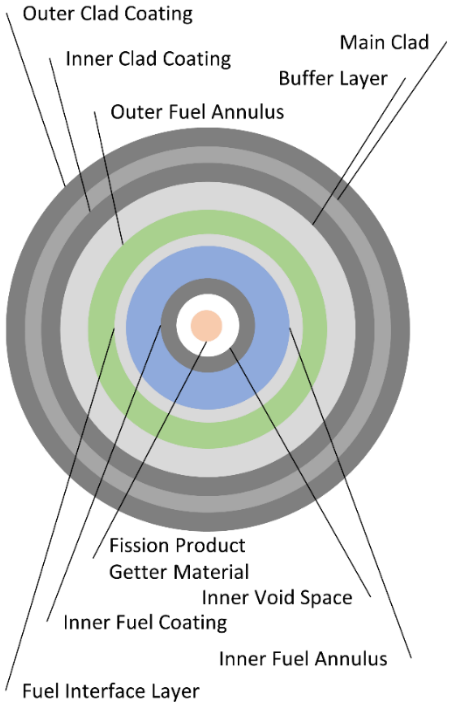


Figure 1. Radial cross section view of fuel element geometry (not to scale).

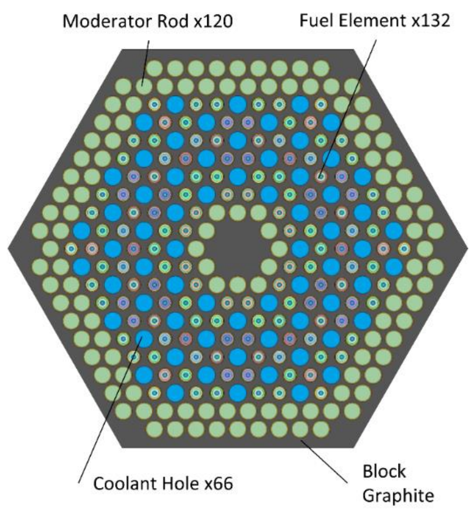


Figure 2. Prismatic block with annular fuel.

TABLE I. Annular Fuel Pin Regions

Layer	Material	Outer Radius (cm)	Inner Radius (cm)
Outer Clad Coating	Pyrolytic Carbon	0.625	0.621
Main Clad	Silicon Carbide	0.621	0.561
Inner Clad Coating	Pyrolytic Carbon	0.561	0.557
Buffer Layer	Carbon Buffer	0.557	0.547
Outer Fuel Annulus	(Pu+Th)CO	0.547	0.417
Fuel Interface Layer	Carbon Buffer	0.417	0.413
Inner Fuel Annulus	(Pu,Th)CO, or ThCO, or (DU,Th)CO	0.413	0.213
Inner Fuel Coating	Pyrolytic Carbon	0.213	0.209
Inner Void Space	Vacuum	0.209	0.109
Fission Product Getter Material	Porous Graphite	0.109	0.000

TABLE II. Pu+Th Configuration

Configuration	Description
10Pu+Th	10 wt% Pu + 90 wt% Th in both annuli (homogeneous)
20Pu+Th | Th	Outer annulus: 20 wt% Pu + 80 wt% Th Inner annulus: 100 wt% Th
20Pu+Th | DU+Th	Outer annulus: 20 wt% Pu + 80 wt% Th Inner annulus: 10 wt% DU + 90 wt% Th
20Pu+Th	20 wt% Pu + 80 wt% Th in both annuli (homogeneous)

3. CALCULATION METHODOLOGY

The lattice physics calculations are performed using the SERPENT 2 (version 2.1.31) Monte Carlo (MC) neutron transport and burnup/depletion code [9]. All results presented in this paper are calculated using the ENDF/B-VII.0 nuclear data library that is distributed with SERPENT 2.
All SERPENT calculations of the lattice physics model of a single prismatic fuel block are conducted using reflective boundary conditions on a fuel assembly. In order to calculate the effective neutron multiplication factor (k-effective, or k_{eff}) taking into account expected neutron leakage in a full, finite-sized reactor core, a 2-group diffusion leakage model with homogenized cross-sections generated by SERPENT 2 is used along with a user-defined geometric buckling value associated with the full finite core geometry of the MHTGR-350 design concept. The assumed value of geometric buckling is ~2.61e-4 cm^{-2}, with a bare core height of ~793 cm (10 blocks per fuel column) and an effective bare core radius of ~153.5 cm (based the area of 66 fuel columns in the MHTGR-350 core), and an assumed zero extrapolation distance. This value of buckling is considered to be conservative, in that it neglects the impact of axial and radial reflectors in reducing neutron leakage. The formula for calculating k_{eff} is given in Equation (1). This calculation provides an approximate value of k_{eff} for comparison purposes in this study. The effects that reflectors have on neutron flux and leakage will likely differ substantially between fuel concepts with and without ^{7}LiH moderator, thus future studies will involve analyzing these concepts in a full core physics model to better quantify their differences in performance.

$$k_{eff} = \frac{\nu\Sigma_{f1} + \nu\Sigma_{f2}\dfrac{\Sigma_{S(1\rightarrow 2)}}{(D_2B^2 + \Sigma_{R2})}}{(D_1B^2 + \Sigma_{R1}) - \Sigma_{S(2\rightarrow 1)}\dfrac{\Sigma_{S(1\rightarrow 2)}}{(D_2B^2 + \Sigma_{R2})}} \tag{1}$$

The single-batch exit burnup and fuel residence time correspond to the burnup step in which $k_{eff} = 1.000$. A two-point linear interpolation is used to estimate the burnup and fuel residence time that correspond to $k_{eff} = 1.000$ using the values of k_{eff}, burnup, and fuel residence time at the last burnup step where $k_{eff} > 1.000$ and at the first burnup step where $k_{eff} < 1.000$. In this study a 3-batch refueling scheme is used. The linear reactivity model is used to estimate the exit burnup and fuel residence time for a 3-batch refueling scheme, which is 3/2 times the single-batch exit burnup and fuel residence time, respectively.

Fissile utilization is a measure of the energy produced per initial mass of fissile material in the fuel. It is calculated as the exit burnup divided by the wt% of fissile material in the fuel at beginning of cycle (BOC). Fissile utilization is a metric that allows comparison between fuels with different initial fissile loadings.

4. RESULTS

Results of the depletion calculations indicate that burnup and fuel residence time are mostly affected by the initial fissile content, as shown in Figure 3 and Figure 4, respectively. They also show that the burnup is ~35 to 45 MWd/kgHM (10 to 15 years) with ~7 wt% fissile and 71 to 95 MWd/kgHM (25 to 35 years) with ~14 wt% fissile. For comparison, the benchmark MHTGR fuel assembly with uranium enriched to 15.5 wt.% $^{235}U/U$ has a predicted exit burnup of 80.7 MWd/kgU, fuel residence time of 3.2 years and fissile utilization of 520.8 MWd/kg-fiss [4]. These burnup values are lower than one might initially expect for fuels with such high fissile content, particularly if they were used in a PWR. However, the small HTGR core, even with a hydrogen-based moderator, and with a geometric buckling of ~2.61e-4 cm^{-2}, experiences a higher level of neutron leakage (40 mk to 50 mk) than that of larger PWR cores (30 mk to 35 mk); (1 mk = 100 pcm = 0.001 $\Delta k/k$). The 10Pu+Th and 20Pu+Th|Th results show that varying the Pu content between the inner and outer annuli has relatively little effect. Replacing 10 wt% of the Th in the inner annulus with

DU results in a 5% decrease in burnup and fuel residence time, which may be due to the higher resonance neutron absorption in U-238 relative to Th-232.

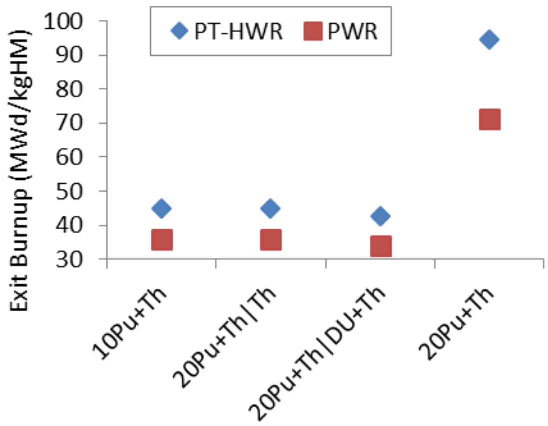
Figure 5 shows that performance with respect to fissile utilization differs between fuel with Pu from a PWR and PT-HWR. With PWR plutonium, the fissile utilization with 10 wt% Pu is slightly higher than that with 20 wt% Pu, whereas it is highest with 20 wt% PT-HWR Pu. For both types of Pu, the configurations with DU have the lowest fissile utilization, due to their lower burnup.

The proportion of Pu found in spent fuel (SF) is shown in Figure 6. Typically, there is 5.5 wt% to 6.5 wt% Pu/IHM in spent fuel, except for the 20 wt% Pu case, where there is 11 to 12 wt% Pu/IHM. In the cases with Pu from a PT-HWR and a PWR, without DU, 54% to 58% and 60% to 63% of the initial Pu remains in SF, respectively. The U-238 in 20Pu+Th|DU+Th fuel leads to higher content in SF. The presence of DU in fresh fuel also has a substantial effect on the fissile quality of plutonium in SF, as is shown in Figure 7. This effect is due to slightly lower burnup with DU, and to breeding of plutonium from U-238. This breeding results in the inner annulus SF comprising ~0.3 wt% Pu/IHM, which is 77 wt% to 83 wt% fissile/Pu. The fissile quality of Pu in SF also depends on burnup and on its quality in fresh fuel:

- higher burnup of 20Pu+Th results in lower fissile quality relative to 10Pu+Th, and
- cases with higher-quality Pu from PT-HWRs result in higher fissile quality in SF, despite their higher burnup.

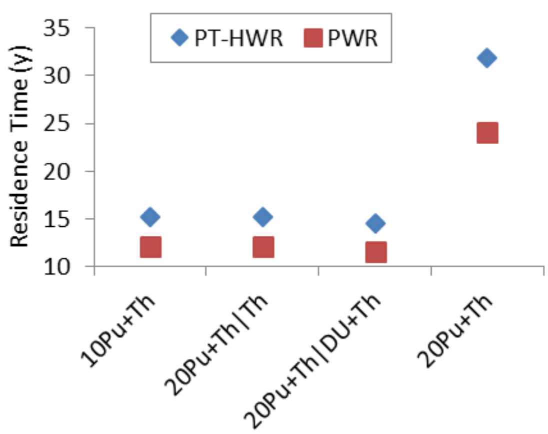
The minor actinide (MA) content in SF depends on the quantity and fissile quality of Pu in fresh fuel, as is shown in Figure 8. The MAs (mainly isotopes of Am and Cm) are produced primarily from neutron capture on Pu isotopes, especially Pu-240 and Pu-242. Plutonium from a PT-HWR has a lower fraction of Pu-240 and Pu-242 than that from a PWR, which results in lower MA production.

The presence of Th in the fuel leads to the production of U-233. Figure 9 shows that the SF is 0.8 wt% to 1.5 wt% U-233/IHM, and that higher U-233 production occurs with higher burnup. The Pu-Th fuels using PT-HWR Pu achieve a higher production of U-233, due to reduced parasitic neutron absorption in the smaller inventories of Pu-238, Pu-240, and Pu-242. Using the Pu content and quality data shown in Figure 6 and Figure 7, along with the U-233 content data shown in Figure 9, the estimated fissile content in the spent Pu-Th fuel ranges between 3 and 6.7 wt% fissile/IHM, with the lowest values found for fuels using PWR-based plutonium. This fissile content is high enough such that the spent HTGR-SMR could potentially be recycled directly, without fissile fuel addition, and with minimal reprocessing, as fuel for a large-scale PT-HWR, or perhaps even a large-scale PWR. Previous studies of Pu-Th fuels in PT-HWRs [10]-[17] have demonstrated that good levels of burnup (≥30 MWd/kg) can be achieved with a fissile content of 2 wt% Pu-fissile/(Pu+Th).



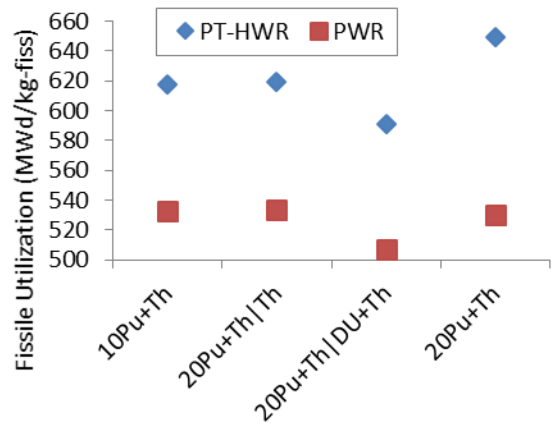
*Exit burnup of benchmark MHTGR TRISO fuel assembly = 80.7 MWd/kgU

Figure 3. Predicted exit burnup for HTGR with plutonium from PWR and PT-HWR spent fuel.



*Fuel residence time of benchmark MHTGR TRISO fuel assembly = 3.2 years

Figure 4. Predicted fuel residence time for HTGR with plutonium from PWR and PT-HWR spent fuel.



*Fissile utilization of benchmark MHTGR TRISO fuel assembly = 520.8 MWd/kg-fiss

Figure 5. Predicted fissile utilization for HTGR with plutonium from PWR and PT-HWR spent fuel.

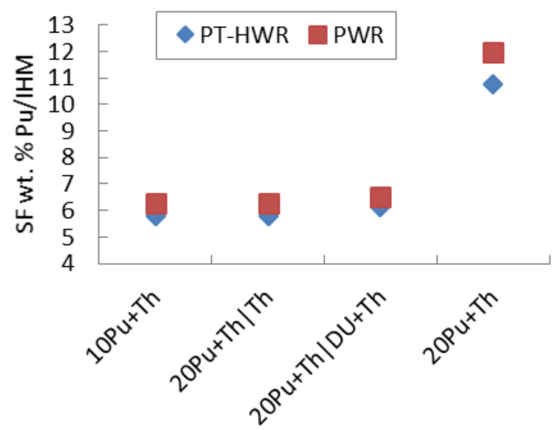


Figure 6. Predicted plutonium content in spent HTGR fuel with initial Pu from PWR and PT-HWR spent fuel.

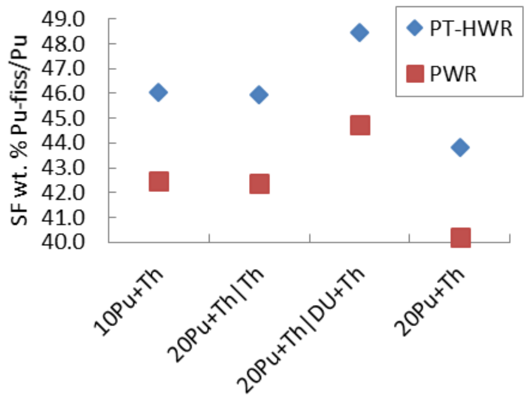


Figure 7. Proportion Pu-239+Pu-241 in spent fuel plutonium

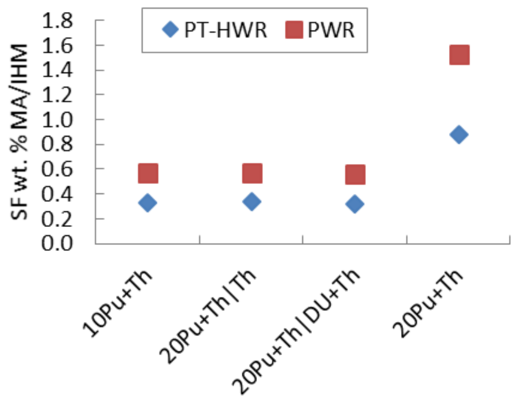


Figure 8. Minor actinides in spent fuel

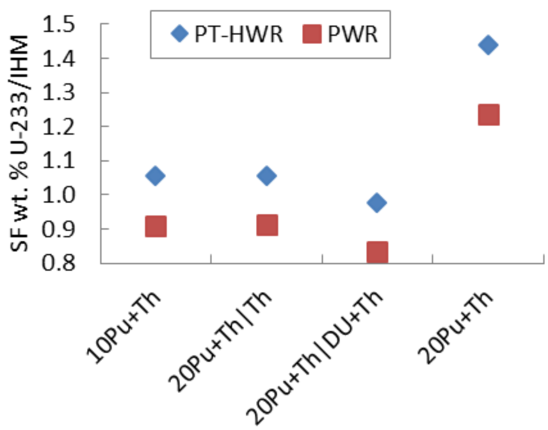


Figure 9. U-233 in spent fuel

5. SUMMARY AND CONCLUSIONS

Lattice physics models and results are presented on burning homogeneous and heterogeneous plutonium-thorium (Pu,Th) and (Pu,Th+DU) fuels in a HTGR using an annular fuel pellet concept in prismatic fuel blocks, with neutron moderation enhanced by the use of hydrogen-based moderator rods made of ^{7}LiH. Results indicate that fuel made of (10 wt% Pu + 90 wt% Th) can achieve burnups of 34 to 44 MWd/kgIHM, while fuel made of (20 wt% Pu+80 wt% Th) can achieve burnups of 71 to 94 MWd/kgIHM, depending on the type of plutonium used for (Pu,Th) and (Pu,Th+DU), and other factors. The high heavy metal loading density of fuel permitted by the use of an annular fuel pellet concept instead of TRISO particle fuel results in fuel residence times greater than 10 years. Thus, the fuel concepts studied could be an attractive option for HTGR-SMRs to achieve compact, long-lived cores. In addition, the use of (Pu,Th) represents a fuel supply option that is an alternative to using high-assay low enriched uranium (HALEU, ~19.75 wt% U-235/U).

Results also show that up to 46% of the plutonium in fresh fuel can be burned in a single burnup cycle, with the discharged plutonium having a lower fissile content (40% to 46% Pu-fissile/Pu). The fissile uranium bred from the thorium (mainly U-233, and trace amounts of U-235) is potentially attractive for recycling, although it makes up less than 1.4 wt% of the spent fuel. The use of thorium instead of depleted uranium (containing U-238) as the main fertile fuel to mix with plutonium also helps limit the production of minor actinides (isotopes of Np, Am, Cm, etc.) to less than 1.6 wt% of the spent fuel. Even when small amounts of depleted uranium are mixed in with the thorium to help denature the U-233, the production of minor actinides is not affected substantially.

It is anticipated that the spent (Pu,Th) and (Pu,Th+DU) annular fuel from HTGR-SMRs could be recycled for subsequent use in a larger scale fast spectrum reactor, or perhaps in a large-scale PT-HWR with high neutron economy. Given that the fissile content in the spent (Pu,Th) fuel, including the Pu-239, Pu-241, and U-233, is at least 3 wt% fissile/IHM, there is more than sufficient fissile fuel available to achieve good burnup levels (>30 MWd/kg) in a PT-HWR [10 to 17]. Future studies can be carried to evaluate the performance capabilities of such fuel cycle options.

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