HTGR technology family assessment for a range of fuel cycle missions

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Abstract

This paper reviews how the high-temperature gas reactor (HTGR) technology family can provide options for the once-through, single recycle or sustained recycle fuel cycle strategies. The HTGR can serve all the fuel cycle missions that a light water reactor (LWR) can; both are thermal reactors. Additional analyses are warranted to determine if HTGR "sustained recycle" service could provide improved consumption of transuranic (TRU) material than LWR can (as is expected), to analyse the unique proliferation resistance issues associated with the "pebble bed" approach, and to further test and analyse methods to separate the tristructural isotropic (TRISO)-coated fuel particles from graphite and/or to separate used HTGR fuel meat from its TRISO coating. The feasibility of these two separation issues is not in doubt, but further R&D could clarify and reduce the cost and enable options not adequately explored at present. The analyses here and the now-demonstrated higher fuel burn-up tests (completed after the illustrative designs studied here) should enable future single-recycle and sustained-recycle HTGR concepts to more rapidly consume TRU, thereby offering waste management advantages. Interest in "limited separation" or "minimum fuel treatment" separation approaches motivates study of impurity-tolerant fuel fabrication.

Introduction

This paper is a summary of a study [1] performed to fill in some of the knowledge gaps for the high-temperature gas reactor (HTGR) technology family with regard to the range of possible fuel cycle approaches. The objective is to examine this technology family in the sense of the Option Study [2]. This helps identify key issues, unknowns and uncertainties. This study primarily addresses the breadth of possibilities and harvests past work.

Several issues are outside the scope of this study, including the following: thorium fuel cycles, gas-cooled fast reactors, reliability of tristructural isotropic (TRISO)-coated particles (billions of such particles in a reactor), and how soon any new reactor or fuel type could be licensed and then deployed and therefore impact fuel cycle performance measures.

HTGR in the context of LWR

As the HTGR and light water reactor (LWR) are both thermal neutron spectrum reactors, the report frequently compares the two as suggested by authors of the Option Study [2]. There are four major LWR-HTGR differences with fuel cycle implications. Compared to LWR, HTGR have a different (and solid) moderator, higher operating temperatures, higher fuel burn-up associated with the TRISO fuel coating, and the "pebble bed" design approach (as opposed to the "prismatic" design approach, which is more directly comparable to LWR).

The solid moderator has several effects. First, it means that there is no reactivity feedback accident sequence involving voiding of the core's moderator. Thus, there is no void coefficient problem in HTGR. The void coefficient issue in LWR constrains TRU loading in fuel recycled to LWR [3]. The lack of the void coefficient problem in HTGR suggests that HTGR could consume transuranics in recycled fuel faster than LWR could. Second, carbon is a less effective moderator than hydrogen (or deuterium), leading to neutron energy spectral changes that slightly decrease uranium utilisation relative to an LWR¹. This reactor physics disadvantage is compensated by the higher operating temperatures, hence higher thermal efficiency. Third, the solid moderator in an HTGR (graphite) represents more of a waste management issue than the liquid moderator in an LWR (water). Heavy metal is 57% (BWR) to 70% (PWR) of the mass of LWR UOX assemblies, i.e. the discharged mass to be managed from used fuel is ~1.5 times that of the heavy metal. But, in the illustrative HTGR designs considered here, which are not optimised, the discharged mass to be managed is ~19 times that of the heavy metal. (Fast reactor designs are intermediate between LWR and HTGR in this regard, with discharged fuel assemblies being 3 to 12 times that of heavy metal.)

The higher fuel burn-up (~10% in the illustrative designs for UO_2 versus ~19% in recent tests for UO_2 fuel) decreases the mass of used fuel (when normalised to the energy produced) that must be disposed if it is not recycled. Higher burn-up slightly decreases radiotoxicity (when normalised to the energy produced) in single recycle and sustained recycle cases. A single recycle in either LWR or HTGR cases reduces radiotoxicity very little in uranium-TRU fuels (MOX) and two to four times in all-TRU fuels (inert matrix fuels, also known as "deep burn"). Sustained recycle of all transuranics in any reactor reduces long-term radiotoxicity by ~2 orders of magnitude, assuming process loss rates below 1%. For sustained recycle cases, the reduction of radiotoxicity in waste depends not so much on the reactor, but rather on the number of times material is recycled and what the processing loss rate would be. The transuranic consumption rate to attain such order-of-magnitude reduction is improved as the TRU loading (TRU per uranium) increases, which is constrained by void coefficient in LWR but not in HTGR.

The pebble bed design approach is outside common safeguard experience. LWR, fast reactors, and prismatic HTGR have fuel assembly masses in the range of 120 to 660 kg and so the impact of the mass of the fuel assembly on the ability to safeguard that assembly is in the same

^{1.} The typically higher burn-up of current HTGR concepts is not enabled by higher in situ conversion of ²³⁸U to ²³⁸Pu, rather there is higher initial ²³⁵U enrichment.

order of magnitude; but the fuel pebbles used in HTGR pebble bed designs each weigh only about 0.2 kg, which could differently impact the fuel pebble safeguardability. (Each pebble has ~50 000 TRISO-coated fuel particles.) Thus, LWR, fast reactors and prismatic HTGR have hundreds of fuel assemblies, which are easily counted as individual items. Pebble bed HTGR have hundreds of thousands of fuel pebbles, e.g. the Pebble Bed Modular Reactor (PBMR)-400 design had 450 000 pebbles. These are thought to be counted in safeguard accounting as bulk material, not individual items. Moreover, the packing density of pebbles varies, making exact balance of the number of pebbles difficult; fortunately, several thousand pebbles are required to make up one "significant quantity" (SQ) of weapons-usable material.

Definitions and terminology

We considered three fuel cycle strategies:

- Once-through any option that discards used fuel without any recycling or post-irradiation processing.
- Single recycle any option that recycles used fuel once. Used fuel from the single recycle is then disposed without any further recycling or post irradiation processing.
- Sustained recycle any option that sustainably recycles used fuel. Used fuel is never directly disposed; one or more TRU elements are always recovered and recycled.

Recycled fuel contains the full range of TRU elements (all-TRU) unless otherwise stated, in which case the fuel is denoted as Pu, NpPu or NpPuAm. In those cases, the un-recycled TRU elements are discarded in waste streams.

Recycled fuel falls into three categories. Mixtures of uranium and one or more TRU elements is called mixed-oxide (MOX) or analogues thereof. Fuel without uranium is called inert matrix fuel (IMF) denoting the fact that the matrix holding the fissile fuel together is inert, neither fertile nor fissile. Fuel with a heterogeneous mixture of UOX and IMF fuels is designated UOX&IMF.

A class of once-through fast reactor concepts is currently being called "breed and burn". These follow the once-through fuel strategy, fresh fuel is enriched uranium; the intent is to discard used fuel. Except for a reactor with 100% enriched ²³⁵U fresh fuel, all reactors breed some ²³⁹Pu (from ²³⁸U) or ²³³U (from ²³²Th) and burn some of it *in situ*. Thus, all reactors are, strictly speaking, breed and burn.

The phrase "deep burn" is used two ways in the literature – high burn-up of uranium fuel or high burn-up of uranium-free fuel. In this report, it is only used in the latter way. For similarity with LWR uranium-free fuel, in figures and tables, the common designator of IMF is used, e.g. LWR-IMF and HTGR-IMF.

By definition, the HTGR family is limited to high-temperature gas coolant reactors and therefore has potential applicability to both electricity and process heat markets. Therefore, apart from fuel cycle considerations, the HTGR may be deployed for process heat with LWR maintaining their electricity market, or the HTGR could also displace the LWR for electricity.

This study follows the definition of the Generation IV programme [4]: HTGR have a thermal neutron energy spectrum. This study does not address the related gas-cooled fast reactor (GCFR), except to note where a few GCFR options border with HTGR options. The only potential fuel cycle mission that cannot be met by the HTGR (but can be by the GCFR) is fuel breeding so that all of the original uranium ore (or thorium in the case of those cycles) is eventually fissioned.

Option space

This study is limited to the U-Pu set of fuel cycles. It does not address thorium fuel cycles.

The study discusses which HTGR characteristics are inherent. Fundamental considerations lead to the conclusion that carbon is the obvious moderator regardless of fuel cycle mission.

However, the HTGR technology family is not limited to TRISO-coated fuel when considering potential application to the range of fuel cycle strategies. And, the HTGR technology family is also not limited to the common separation technology assumption of taking aqueous separation technologies developed for LWR oxide fuels and adding a step of mechanically breaking TRISO fuel coatings and applying LWR techniques to the fuel meat. Figure 1 illustrates the range of fuel and fuel separation options.

- In once-through cases when there is no separation, the logical fuel choice is TRISO-coated particles.
- If used HTGR fuel is to be recycled then the fuel kernels must be separated from the coatings; there are a range of options from crushing to powder to burning. Most of these would be followed by dissolution of the fuel exposed by the initial treatment. A range of fuel options exist: more robust options (such as TRISO) may be harder to disassemble.
- In "limited" separations cases such as only heating to release gaseous fission products, the most robust fuel options (TRISO) do not apply.



Figure 1: Range of HTGR fuel and separation options

Key results and considerations

The HTGR is considered for deployment independent of fuel cycle objectives; HTGR fuel cycle analyses are therefore appropriate regardless of perceived fuel cycle performance – to know how HTGR deployment would impact the overall fuel cycle prospects and options and what HTGR-relevant technologies should be researched. If substantial HTGR deployments occur, there is little reason at present to believe that that would worsen the prospects for advanced fuel cycles. Depending on the fuel cycle mission, those technologies could include new fuels with their fabrication techniques, new fuel separation approaches – either "full" separation or "minimum fuel treatment" – or new reactor designs.

To enhance reactor safety and reduce operational radiation fields around coolant equipment, the mainline HTGR approach is to develop a very robust fuel coating (TRISO). The HTGR potential for a very low radioactive coolant is unparalleled as helium (or CO₂) is non-radioactive and is less corrosive/erosive than water, sodium, lead, molten salts, etc. [5]. Thus, the value of an extremely robust coating (with a very small fission product escape probability into the coolant) is higher in an HTGR than for other reactor coolants and thus a long-standing objective of HTGR R&D is creation of very robust fuel coatings and assembly into structures that face the helium coolant. However, for recycle strategies, a robust fuel coating. Options for undoing TRISO coatings that have received little attention include jet-milling followed by mechanical re-forming, AIROX or traditional chemical separation. Options for less robust coatings that have received little attention include jet.

When recycling HTGR fuels, the coated fuel particles must first be separated from the pebbles (in pebble bed designs) or from the compacts (in prismatic designs). Options for separating particles from the pebbles or compacts include mechanical separations and passing electric currents through the fuel, which can disintegrate the fuel form leaving matrix power and fuel particles.

The relatively poor coolant characteristics of gas, the single-phase nature of such coolants, and the desire for high thermal efficiency have led gas reactors to high temperatures, embedded in the names HTGR and VHTR. The high operating temperatures may offer a way to release fission products during operation, an idea being studied in General Atomics' EM² concept. Although EM2 is a once-through fast reactor concept starting with plutonium fuel, the approach has potential applicability to uranium-fuelled thermal reactors as well; indeed removal of neutron absorbing fission products would have more value in thermal reactors than fast reactors.

The FY2009 losses study report [6] noted the linkage between impurities of waste in fuel products and impurities of fuel materials in waste products (e.g. TRU in waste). Higher tolerance of impurities in fuel may offer less TRU impurities in waste and conceivably even eliminate HLW altogether when defined by its characteristic components, studied in the United States, Japan, South Korea and California [7-10]. (Of course, under current United States law, all fission products could be HLW independent of their waste characteristics.) Meanwhile, to potentially reduce proliferation concerns and cost, DOE wants study of "minimal fuel treatment" options in addition to traditional full-separation techniques. This creates recycle fuels with high impurities. It is not clear how the potential of HTGR to tolerate high-impurity recycle material differs from other technology families.

All commercial reactors in the United States and 85% of those in the world are LWR. About 10% of the reactors around the world are heavy water, 4% are gas-cooled and <0.5% are liquid-metal-cooled. Any non-LWR reactor's contribution to advanced fuel cycle strategies is constrained by how fast and how much the new reactor type is deployed.

For example, consider a single-recycle (deep burn) scenario in which all HTGR are deep burn, using TRU from used LWR UOX fuel. On a GWth basis, the HTGR would have to be 13% of the combined LWR-HTGR fleet. On a GWe basis, this is about 10%. The United States has 104 operating LWR, producing about 2 000 tonnes of used fuel per year while generating about 90 GWe-year of electricity per year. To balance that, one would need 2.5 modules of 800 tonne-UOX/year separation units and 9 GWe-year electricity from HTGR. At 90% capacity factor, this would require about 10 GWe capacity of HTGR, or more than two dozen PBMR-400.

Waste

Figure 2 shows the radiotoxicity of waste for several LWR and HTGR cases. The three black lines are once-through cases with similar burn-ups (100 to 110 MWth-day/kg-iHM); there is little difference among them. The uranium-free single recycle cases for both HTGR and LWR are uranium-free IMF cases (called deep burn in HTGR); they show significant radiotoxicity reduction between 100 and 100 000 years after reactor discharge primarily due to consumption of ²³⁹Pu;



Figure 2: Radiotoxicity for LWR and HTGR options

Solid lines are LWR, dashed lines are HTGR prismatic, dotted lines are HTGR pebble bed

there is an increase of radiotoxicity between 100 000 and 1 000 000 years due to production of some of the fertile isotopes. Single recycle MOX fuels in LWR and HTGR (not shown) exhibit little or no radiotoxicity reduction versus once-through. The sustained recycle of LWR fuel reduces long-term radiotoxicity by orders of magnitude versus both once-through and single recycle LWR and HTGR cases; this would be expected to occur with sustained recycle HTGR as well but there are no known analyses of that option.

More generally, there are two ways to achieve orders of magnitude of radiotoxicity reduction relative to once-through: sustained recycle of all TRU (with less than 1% TRU loss to waste) and/or an external supply of neutrons such as from a fission fusion hybrid to burn 99% of the initial heavy metal.

Table 1 helps illustrate the potential importance of separating fuel meat (kg-iHM/assembly) from other in-core material (kg-total/assembly). The ratio of total mass to fuel meat mass varies from 1.4 to 106. Even if the fuel meat (uranium, TRU) is not to be recycled, there can be significant reduction in total mass and volume if the non-fuel materials can be separated from the fuel meat.

Uranium utilisation

Figure 3 shows uranium utilisation for LWR and HTGR concepts. Uranium utilisation in both LWR and HTGR is improved with recycling, but still remains below 1%. ~100% utilisation can only be achieved by full recycle with fast breeder reactors. Recycling in thermal reactors improves uranium utilisation, but only modestly. The two HTGR-UO2-prismatic cases have unusually low uranium utilisation; this results from neutron spectral differences versus LWR-UOX. The HTGR-UO2-pebble bed case does not appear to suffer as much, presumably because of the efficiencies associated with constant shuffling of fuel.

	kg-iHM/	kg-total/	Ratio of total
	assembly	assembly	mass to IHM
Once through PWR UO ₂	461.3	657.9	1.4
Single or sustained recycle LWR MOX	461.3	657.9	1.4
Single or sustained recycle LWR IMF-Pu	37.2	530.9	14.3
Single or sustained recycle LWR IMF-NpPuAm	42.7	580.8	13.6
Single or sustained recycle LWR IMF-TRU	42.5	578.8	13.6
Once-through BWR UO ₂	183.3	319.9	1.7
	kg-iHM/ assembly	kg-total/ assembly	Ratio of total mass to iHM
Sustained recycle FR metal CR = 0.00	29.7	349.7	11.8
Sustained recycle FR metal CR = 0.25	44.9	342.8	7.6
Sustained recycle FR metal CR = 0.50	69.9	366.1	5.2
Sustained recycle FR metal CR = 0.75	97.7	411.5	4.2
Sustained recycle FR metal CR = 1.00	114.1	446.1	3.9
Sustained recycle FR oxide CR = 0.00	37.7	264.9	7.0
Sustained recycle FR oxide CR = 0.25	60.6	335.6	5.5
Sustained recycle FR oxide CR = 0.50	91.9	426.1	4.6
Sustained recycle FR oxide CR = 0.75	126.0	483.2	3.8
Sustained recycle FR oxide CR = 1.00	148.6	540.0	3.6
	kg-iHM/ element	kg-total/ element	Ratio of total mass to iHM
Once-through HTGR-UO ₂ prismatic	7.2	134.4	18.7
Single recycle HTGR-IMF prismatic	1.2	126.9	105.8
	kg-iHM/ pebble	kg-total/ pebble	Ratio of total mass to iHM
Once-through HTGR-UO ₂ pebble bed	0.011	0.21	19.1
Single recycle HTGR-IMF pebble bed	0.002	0.20	100.0

Table 1: Mass of fuel assemblies in various option
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Figure 3: Uranium utilisation and uranium ore consumption, without credit for thermal efficiency



There is a limitation in the preceding graph; it ignores the differing thermal efficiency among concepts. The Gen-IV roadmap says, "The VHTR can also generate electricity with high efficiency, over 50% at 1 000°C, compared with 47% at 850°C in the GTMHR or PBMR. Co-generation of heat and power makes the VHTR an attractive heat source for large industrial complexes [4]." This report is not limited to any specific HTGR concept or design and therefore it is appropriate to consider how the preceding figure changes with an HTGR thermal efficiency of 50% versus LWR efficiency of 33%.

Figure 4 is identical to Figure 3 except that the normalisation is changed from GW-thermal to GW-electric, i.e. the impact of thermal efficiency is included. In this graph, LWR are assumed to have 33% thermal efficiency and HTGR have an upper-bound value of 50%. CANDU are assumed to a 30% thermal efficiency. Thus, Figure 4 shows a best-case HTGR/LWR comparison. The HTGR-UO2-prismatic case is still somewhat below LWR-UOX. The HTGR-UO2-pebble bed and the HTGR-IMF cases are above the LWR-UOX cases.



Figure 4: Uranium utilisation vs. uranium ore for thermal reactor cases – with thermal efficiencies of 33% for LWR, 50% for HTGR and 30% for CANDU

Proliferation resistance and physical protection

The first key issue is whether fuel fabrication technology would be provided to a host country in addition to the reactor for two reasons.

First, one misuse scenario is making "fake" fuel with ²³⁸U leading to higher quality Pu than routine uranium or recycled Pu or TRU fuel. This Pu would also be "off the books" and thus not safeguarded. Detection of "fake" fuel would be difficult prior to insertion. However, if the host country does not obtain TRISO technology and nonetheless attempts to make fake fuel, it would seem highly likely to fail. Given the high purity and low radioactivity of HTGR coolants, such failures of fake fuel might be detectable.

Second, TRISO fuel fabrication technology involves a degree of separation chemistry to make high chemical purity U (or U-TRU or TRU) feedstock. So, having TRISO fuel fabrication technology would mean a country automatically has a certain chemical separation technology and experience in handling nuclear materials.

With regard to separation of used fuel, an existing and obvious way to recover U or TRU from used TRISO fuel is to burn the pebbles (if pebble bed) or fuel elements (if prismatic). This has two problems. First, it should be very easy to detect off-site. Second, without some crushing, one still has not gotten to the fuel meat, which lies inside both carbon layers (which will burn) and a SiC layer.

The various methodologies consider the smallest unit of mass that can be diverted or stolen. The higher, the better. Tables 4-6 provide the mass of heavy metal and total mass of fuel assemblies (or fuel elements or fuel pebbles). Except for the HTGR pebble bed, all the options have total assembly masses of 120-660 kg. It is unknown whether those are significant differences from a diversion or theft perspective. Even if they were, the values shown in the table were never selected from this perspective. That is, if significant advantage were given to concepts with say 600 kg assemblies versus 100 kg assemblies, perhaps prismatic fuel elements could be increased in size and mass.

The pebble bed approach is an obvious outlier, with 0.2 kg/pebble, each containing only a small mass of heavy metal.

Safeguards are all about counting. What do you have to count? How well? Although not "safeguards" *per se*, similar issues exist for theft scenarios. How much material must you steal? As you transport it elsewhere, how much material must you move and how could that be detected?

The pebble bed design approach is outside common safeguard experience. LWR, fast reactors and prismatic HTGR have fuel assembly masses in the range of 120 to 660 kg; HTGR pebble bed have fuel pebbles about 0.2 kg. (Each pebble has ~50 000 TRISO-coated fuel particles.) Thus, LWR, fast reactors and prismatic HTGR have hundreds of fuel assemblies, which are easily counted as individual items. Pebble bed HTGR have hundreds of thousands of fuel pebbles, e.g. the PBMR-400 design had 450 000 pebbles. These are thought to be counted as bulk material, not individual items. Moreover, the packing density of pebbles varies, making exact balance of the number of pebbles difficult; fortunately, several thousand pebbles are required to make up one "significant quantity" (SQ) of weapons-usable material.

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References

- [1] Piet, Steven J., Samuel E. Bays, Nick R. Soelberg, HTGR Technology Family Assessment for a Range of Fuel Cycle Missions, FCRD-SYSA-2010-000130, 23 August (2010).
- [2] Wigeland, Roald, et al., AFCI Options Study, AFCI-TIO-PMO-MI-DV-2009-000086, Advanced Fuel Cycle Initiative, September (2009).
- [3] Youinou, Gilles, Samuel Bays, Homogeneous Recycling of Pu or Pu+M.A. in PWRs Loaded with MOX-UE Fuel (MOX with ²³⁵U Enriched U Support), INL/EXT-09-16091, AFCI-SYSA-TRAN-SS-RT-2009-000055, June (2009).
- [4] Generation IV Roadmap Technology Goals for Generation IV Nuclear Energy Systems, GIF-019-00, December (2002).
- [5] Smith, D.L., et al., Blanket Comparison and Selection Study Final Report, Argonne National Laboratory Report, ANL/FPP-84-1, September (1984).
- [6] Shropshire, David E., et al., System Losses and Assessment Trade Study, AFCI-SYSA-PMO-MI-DV-2009-000203, INL/EXT-09-16891, 30 September (2009).
- [7] Piet, Steven J., David E. Shropshire, Nick Soelberg, "Must Advanced Fuel Cycles Produce High-Level Waste?" Integrated Radioactive Waste Management in Future Fuel Cycles, Charleston, South Carolina, 8-12 November (2009).
- [8] Arie, Kazuo, et al., "A Strategy on Minimizing High-level Waste Burden for Sustainable Nuclear Energy System", GLOBAL 2009, 6-11 September (2009), paper 9087.
- [9] Nam, Hyo On, et al., "Partitioning and Transmutation System Leaving No High Level Waste Behind: PEACER-PARK", GLOBAL 2009, 6-11 September (2009), paper 9440.
- [10] Fowler, T. Kenneth, Joonhong Ahn, "Toward On-site Closed Nuclear Fuel Cycles Not Requiring Deep Burial of Waste", *Journal of Fusion Energy*, 29 (2), pp. 188-195 (2010).