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A White Paper: Disposition Options for a High-Temperature Gas-Cooled Reactor

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EXECUTIVE SUMMARY

The high-temperature gas-cooled reactor (HTGR) is a uranium-fueled, graphite-moderated, gas-cooled nuclear reactor design concept capable of producing very high core outlet temperatures. Both types of HTGR have the tristructural isotropic (TRISO) fuel kernel at the heart of the fuel design. For the prismatic block-type HTGR, the TRISO particles are overcoated with a resinated graphitic matrix and pressed into fuel compacts, which are then heat treated and placed in the fuel channels of the prismatic-block-shaped fuel assemblies. For the pebble-bed-type HTGR, the TRISO particles are dispersed in a graphitic-matrix sphere, which is the basic unit for the reactor core. Despite having very different fuel designs, both types of HTGR are graphite-moderated, gas-cooled, thermal reactors using many of the same materials. As a result, both prismatic-block-type and pebble-bed-type HTGRs have similar radioactive waste streams, all of which require safe and secure storage and eventual disposition. Modern HTGR designs are based on a long and rich operating history of several different graphite-moderated, gas-cooled, thermal reactors. Several of these reactors have been shut down, the fuel has been placed in safe storage, and they have undergone some degree of decommissioning. As such, there is significant experience in the management of the spent nuclear fuel (SNF) and radioactive wastes associated with operating these reactors.

This white paper will, (1) identify the definitions and regulations that apply to the safe and secure management, storage, and disposal of radioactive waste; and (2) identify the key radioactive waste streams from HTGRs and their characteristics. Idaho National Laboratory (INL) has significant experience in the management of SNF from HTGR predecessors. This experience should form the basis for the management and disposition efforts of the radioactive waste from any new HTGR-type small modular reactor, or microreactor intended for deployment at the INL site.

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ACRONYMS

AEA	Atomic Energy Act of 1954, as amended
AVR	Arbeitsgemeinschaft Versuchsreaktor
CFR	Code of Federal Regulations
DOE	U.S. Department of Energy
FSV	Fort St. Vrain Generating Station
GTCC	Greater-Than-Class C
HLW	High-Level Waste
HTGR	High-Temperature Gas-Cooled Reactor
HTTR	High-Temperature Engineering Test Reactor
INL	Idaho National Laboratory
LLW	Low-Level Waste
NWPA	Nuclear Waste Policy Act of 1982, as amended
PB	Peach Bottom Atomic Power Station
RCRA	Resource Conservation and Recovery Act
SNF	Spent Nuclear Fuel
THTR	Thorium High-Temperature Reactor
VHTR	Very High-Temperature Reactor

1. INTRODUCTION

The high-temperature gas reactor (HTGR) is a uranium fueled, graphite moderated, gas cooled nuclear reactor design concept capable of producing very high core outlet temperatures. There are two types of HTGRs: the prismatic block-type and the pebble bed-type reactors. Both types of HTGRs use tristructural isotropic (TRISO) fuel kernels at the heart of the fuel designs. For the prismatic block-type HTGR, the TRISO particles are overcoated with a resonated graphitic-matrix and pressed into fuel compacts which are then heat treated and placed in the fuel channels of the prismatic block shaped fuel assemblies. For the pebble bed-type HTGR, the TRISO particles are dispersed in a graphitic-matrix sphere, which is the basic unit for the reactor core. Figure 1 shows an overview of how TRISO particle kernels are made into HTGR fuel, for both the prismatic block-type and the pebble bed-type reactors.

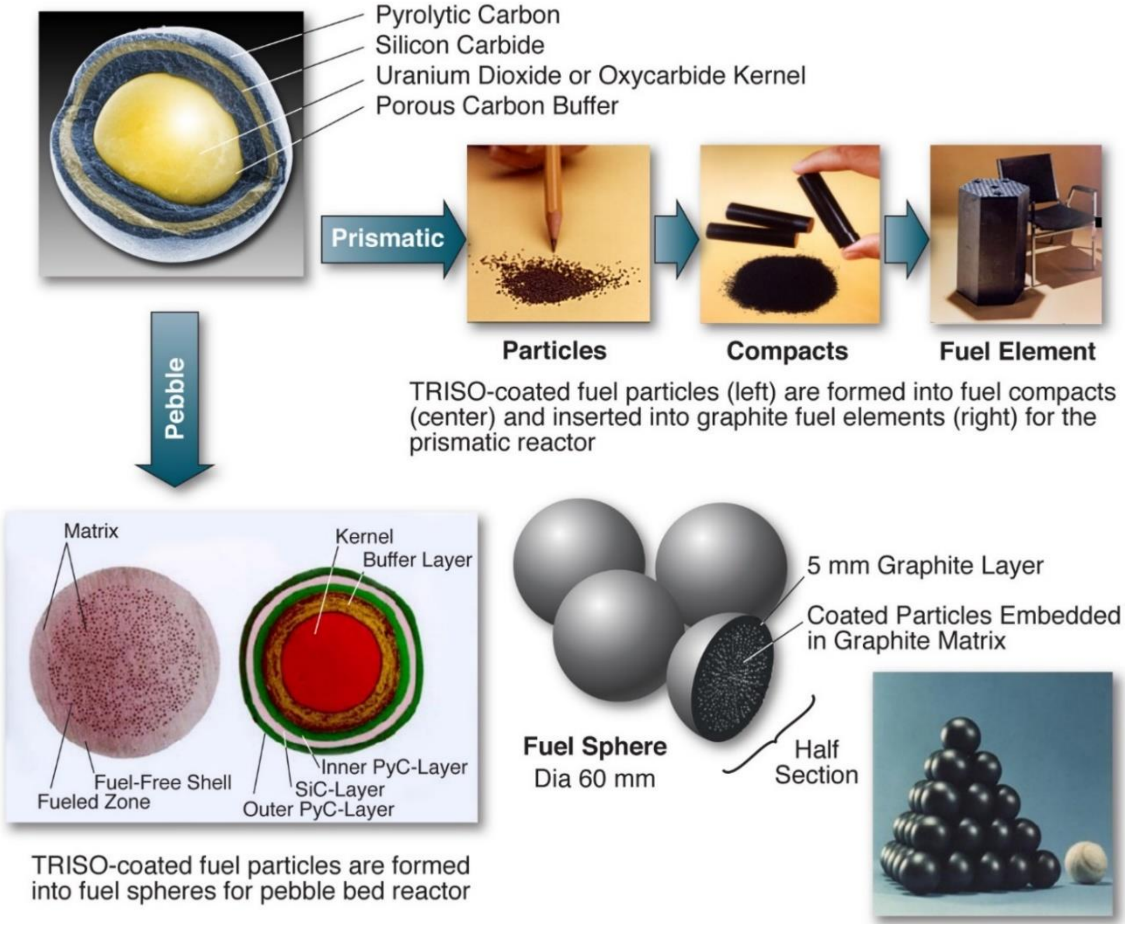

Figure 1. Overview of how TRISO particle kernels are made into both prismatic block-type and pebble bed-type HTGR fuel (Demkowicz et al. 2020).

Modern HTGR designs are based on a long and rich operating history of several helium-cooled, thermal reactors. These include: the Dragon reactor, Peach Bottom Atomic Power Station (PB), Arbeitsgemeinschaft Versuchsreaktor (AVR) reactor, Fort St. Vrain Generating Station (FSV), the Thorium High-Temperature reactor (THTR), the High-Temperature Engineering Test reactor (HTTR), and the HTR-10. The HTR-10 reactor is a prototypic Chinese Very High-Temperature reactor (VHTR) of the pebble bed-type with full-scale reactors coming online soon in China. The VHTR is one of the six, GenIV initiative reactor concepts and is distinguished from the more conventional HTGR by its operating temperature, as a He-cooled reactor capable of outlet coolant temperatures up to 1000°C.

Apart from the HTTR and HTR-10, the remaining HTGR reactors have been shut down, the fuel has been placed in safe storage, and they have undergone some degree of decommissioning. As such, there is significant experience in the management of the spent nuclear fuel (SNF), and radioactive wastes associated with operating these reactors. Other graphite-moderated, gas-cooled reactors, such as the British MAGNOX and Advanced Gas-Cooled Reactor (AGR), also offer relevant and significant experience, even though the coolant used in these reactors was carbon dioxide.

Of the seven HTGR plants constructed and operated, four HTGR plants used prismatic block-type fuel; the remaining three HTGR plants used pebble bed-type fuel. Table 1 presents the location, thermal power output, and years of operation of various prototype and demonstration HTGRs.

Table 1. Overview of prototype and demonstration HTGRs with significant years of operation.

Reactor	Type	Location	Thermal Power (MW)	Years of Operation
Dragon	Prismatic block	Winfrith, England	21.5	1964–1975
PB	Prismatic block	Pennsylvania, United States	115	1966–1974
AVR	Pebble bed	Julich, Germany	46	1967–1988
FSV	Prismatic block	Colorado, United States	842	1976–1989
THTR	Pebble bed	Hamm Uentro, Germany	750	1985–1991
HTTR	Prismatic block	Tokaimura, Japan	30	1998–present
HTR-10	Pebble bed	Beijing, China	10	2000–present

While having very different fuel designs, both types of HTGR (prismatic block-type and pebble bed-type) are graphite-moderated, gas-cooled, thermal reactors using many of the same materials. As a result, both prismatic-block-type and pebble-bed-type HTGRs have similar radioactive waste streams, all of which require safe and secure storage, and eventual disposition. This includes the SNF, in core graphite, and stainless-steel components, piping, resins and filters, solidified liquid waste, contaminated equipment, and other radioactive wastes.

Section 2 discusses the classification of radioactive waste and how these classifications may apply to HTGR waste streams. Section 3 identifies the key classes of HTGR radioactive waste streams. Idaho National Laboratory has significant experience in the management of SNF from the HTGR predecessors; specifically, the FSV SNF, which has 23 metric tons stored, and PB SNF, which has 3 metric tons stored. This INL experience should form the basis for the management and disposition efforts of the radioactive waste from any new HTGR-type, small, modular reactor or microreactor intended for deployment at the INL site. Section 4 presents the conclusions of this paper.

2. RADIOACTIVE WASTE CLASSIFICATION

The disposition options for radioactive waste streams from HTGR operations will be dictated, by the classification of the radioactive waste streams, as either SNF, high-level waste (HLW), low-level waste (LLW), transuranic (TRU) waste, or as byproduct material. The applicable definitions for the various categories of radioactive waste in the U.S. are established by the U.S. Department of Energy (DOE), the Atomic Energy Act of 1954 (AEA) (Atomic 1954), and the Nuclear Waste Policy Act of 1982 (NWPA) (Nuclear 1982). The following are the terms as defined in the relevant laws.

- **Spent nuclear fuel (NWPA 1982)** – fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing.
- **High-level waste (NWPA 1982)** – (A) the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and (B) other highly radioactive material that the Commission, consistent with existing law, determines by rule requires permanent isolation.
- **Transuranic waste (DOE Order 435)** – radioactive waste containing more than 100 nanocuries (3700 becquerels) of alpha-emitting transuranic isotopes per gram of waste, with half-lives greater than 20 years, except for: (1) High-level radioactive waste; (2) Waste that the Secretary of Energy has determined, with the concurrence of the Administrator of the Environmental Protection Agency, does not need the degree of isolation required by the 40 CFR Part 191 disposal regulations; or (3) Waste that the Nuclear Regulatory Commission has approved for disposal on a case-by-case basis in accordance with 10 CFR Part 61.
- **Low-level waste (NWPA 1982)** – radioactive material that (A) is not high-level radioactive waste, spent nuclear fuel, transuranic waste, or by-product material as defined in section 11e(2) of the AEA [42 U.S.C. 2014(e)(2)]; and (B) the Commission, consistent with existing law, classifies as low-level (C) radioactive waste.
- **Byproduct material (AEA 1954)** – (1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing special nuclear material.

There are four classes of LLW: Class A, Class B, Class C, and Greater-Than-Class C (GTCC). These classes are based on waste form requirements and the activity per unit volume of specific radioisotopes, as defined in the Title 10, Part 61 of the Code of Federal Regulations (CFR) (10 CFR 61).

There are four categories of byproduct material: (1) radioactive material that results from the fission, or splitting apart, of enriched uranium or plutonium in nuclear reactors; (2) tailings or waste produced by processing uranium or thorium from ore; (3) certain processed radium-226 or material that becomes radioactive in a particle accelerator used for a commercial, medical, or research activity; and (4) a naturally occurring radioactive source that is processed to increase its concentration, and that the Commission decides could pose a threat to people and the environment similar to that of radium-226.

Depending on the operations at HTGR plants, it is expected that some of the waste streams from HTGRs could be classified as LLW. This has been done for byproduct material, such as components of the primary loop, drainable liquids, resins, filters, solidified liquid waste, contaminated equipment, delayed beds for capturing gases, decontaminated piping, the containment vessel, and refueling equipment, to name a few.

Disposition of some HTGR radioactive waste as either TRU or LLW may be possible following the recent DOE interpretation of HLW (DOE 2018, DOE 2019). DOE interprets that reprocessing waste may be determined to be non-HLW if the waste meets either of the following two criteria: (*1) does not exceed concentration limits for Class C low-level radioactive waste as set out in section 61.55 of title 10, CFR, and meets the performance objectives of a disposal facility; or (2) does not require disposal in a deep geologic repository and meets the performance objectives of a disposal facility as demonstrated through a performance assessment conducted in accordance with applicable requirements*.

Although not expected, any HTGR radioactive waste streams that contain hazardous waste would be classified as mixed waste. Hazardous wastes are materials known, or tested to exhibit one or more of the following hazardous traits: ignitability, reactivity, corrosivity, or toxicity, as per 40 CFR 261. Mixed wastes are regulated by both the Resource Conservation and Recovery Act (RCRA) (Resource 1976) and the AEA.

3. HTGR RADIOACTIVE WASTE STREAMS

The radioactive waste streams from HTGRs can be categorized into the following groups:

1. Spent nuclear fuel
2. Graphite/carbon wastes
3. Other radioactive wastes.

The treatment options for the SNF and graphite waste streams depend on the level to which the prismatic block-type and pebble bed-type fuel are deconstructed. For the prismatic block-type HTGR, the TRISO particles are embedded in fuel compacts, which are then placed in the fuel channels of the prismatic block-shaped fuel assemblies. For the pebble bed-type HTGR, the TRISO particles are dispersed in a graphitic pebble, which is the basic unit for the reactor core. The simplest approach is whole block or whole pebble disposal. There are also increasingly complex options, where these successive layers of graphite structures are removed. This increases the disposal options, but also increases the total volume of radioactive waste. For some HTGR fuel designs, graphite represents up to 95% of the fuel elements (TANG 2002). Reprocessing of HTGR fuel has been studied in the U.S. and in other nations as far back as the 1960s. The primary objective of these studies was the recovery of fissile materials within the TRISO fuel kernel. This required the elimination of the successive layers of carbon and silicon carbide (SiC). After this step, the uranium or thorium (if the thorium fuel cycle was used) were recovered and separated from the fission products by the UREX or THOREX processes. The recovered material could be used to fabricate new fuel and the waste could be discarded. There have been proposals to reuse irradiated graphite in the fabrication of new fuel forms with no consensus on the technical and economic feasibility of such a process.

The radioactive isotopes in irradiated graphite requiring attention are carbon-14 (^{14}C) and tritium (^{3}H). ^{14}C is created from the activation of absorbed nitrogen (^{14}N) and naturally occurring carbon-13 (^{13}C) in graphite, and ^{3}H is generated from the activation of lithium, which is an impurity in graphite.

Other radioactive waste streams include metal, carbon, operating, decontamination, and decommissioning waste streams. In general, these wastes can be treated and disposed of in the same manner as similar wastes from other nuclear reactors.

3.1 Spent Nuclear Fuel

The preferred disposition path for HTGR SNF, depends on the availability and waste acceptance criteria for potential radioactive waste repository. Other factors include the availability and compatibility of the irradiated graphite for disposal as LLW, risk management, and overall cost. Typically, increased SNF processing can decrease the volume of SNF while increasing the volume of LLW. However, increased SNF processing can lead to increased costs and waste management risks. Options for SNF disposal include whole block disposal, prior removal of nonactinide-bearing graphite, separation of actinide-bearing fuel kernel, and recovery of actinide material from fuel kernel. Figure *2* shows the treatment and disposal options for HTGR SNF. The green blocks represent treatment processes. The red blocks represent various waste forms with the likely waste classification in parentheses.

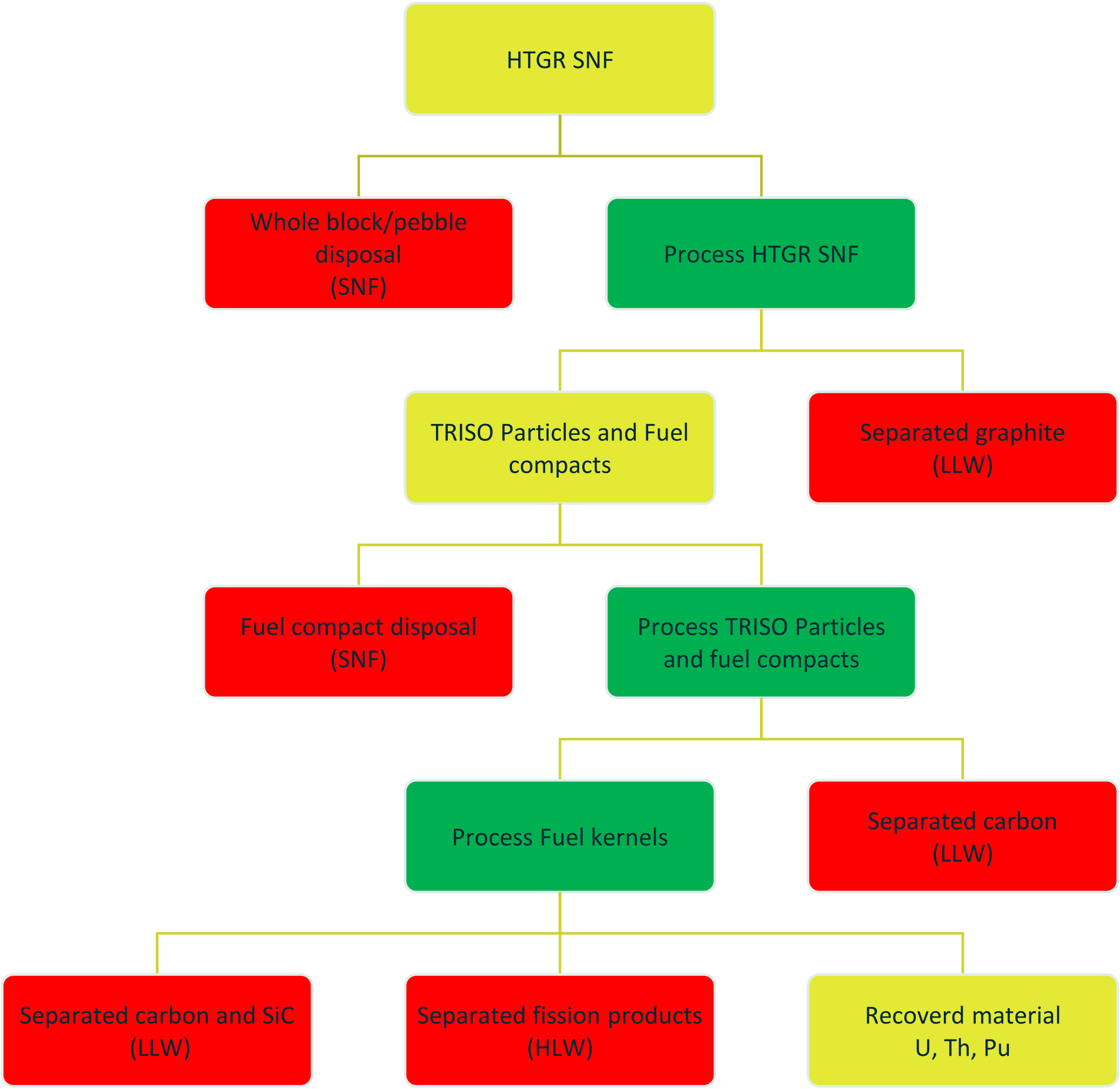

Figure 2. Treatment and disposal options for HTGR SNF.

3.1.1 Whole Block Disposal

Intact and damaged HTGR SNF may be disposed of in repositories if the necessary waste acceptance criteria are met. In this case, the HTGR SNF can be packaged in the appropriate waste canisters. The HTGR SNF typically has a lower decay heat load due to the large volumes of graphite within the SNF. Therefore, HTGR SNF could be candidates for large, more economical waste packages.

3.1.2 Prior Removal of Graphite

If whole block disposal is unacceptable, or material recovery or waste volume reduction is desired, HTGR SNF can be disposed of following the removal of graphite. Various degrees of prior graphite removal can be performed. The simplest is the separation of the SNF matrix from the graphite blocks/sleeve. This can be achieved by grinding the prismatic elements or pebbles, and oxidation of the crushed graphite. Novel approaches involve coring of the SNF matrix to remove the fuel compacts without damaging the fuel kernels with prismatic block HTGR fuels (Greneche et al. 2006). Once the graphite has been removed, the fuel compacts can be packaged for disposal at the repository. The compacts may also be further processed to separate the valuable isotopes from the fission products through oxidation followed by chemical separation. Again, the resulting materials can be appropriately packaged for disposal at the repository. Several overpacking, coating, or encapsulation technologies exist to produce an acceptable waste form.

3.1.3 Dissolution of Spent Fuel

Chemical processing of the HTGR SNF can be achieved via conventional processing techniques that allow for the recovery of the U, Pu, and Th (if the Th fuel cycle was used). The separated fission products can then be encapsulated in glass, or any other appropriate waste form that meets the repository waste acceptance criteria. Electrochemical separation using salt electrolytes has also been investigated as a means to remove the graphite matrix from the HTGR fuels with promising results (Tian et al. 2009). These processes typically leave the SiC coating protecting the inner carbon buffer zone and fuel kernel matrix. The SiC layer can be breached by grinding to expose the inner carbon and fuel kernel. Further treatment methods may then recover the fuel, while leaving the empty SiC shell fragment as waste.

3.2 Graphite/Carbon Waste Streams

The carbon streams from most HTGRs will largely be composed of polycrystalline graphite used as the moderator, reflector, and structural material. The radioactive isotopes in irradiated graphite requiring attention are ^{14}C and ^{3}H. ^{14}C is created from the activation of absorbed ^{14}N and naturally occurring ^{13}C in graphite. ^{3}H is generated from the activation of lithium, which is an impurity in graphite. The activity levels of ^{14}C in some cases may be greater than 8 Ci/m^3 limit for Class C LLW as stated in Title 10, Part 61 of the CFR (10 CFR 61). Technologies to reduce the amount of absorbed ^{14}N in the graphite during manufacture, such as inert nonnitrogen-bearing atmospheres (like argon), could significantly reduce the nitrogen impurity. ^{3}H generation is estimated to be significantly less than the 40 Ci/m^3 limit for Class A LLW.

The irradiated graphite has significant value for recycle and reuse. Not only does this reuse a valuable material but also greatly decreases the volume of waste to be disposed of. In general, there are three options for graphite disposal: (1) direct disposal without any chemical treatment, (2) graphite gasification, and (3) decontamination and recycling. Graphite gasification can be achieved through oxidation in oxygen or gasification with steam. Direct disposal of graphite in its original form has the advantages of requiring no additional processing and being a natural trap for the radioactive ^{14}C to the environment, but has the disadvantage of large waste volume. Graphite gasification has the advantage of large repository waste volume minimization, but with the disadvantage of generation of greenhouse gases (CO_2), requiring capture technology. In addition, ^{14}C and ^{3}H become CO, CO_2, and H_2O along with the rest of the graphite and carbon. This results in a radioactive gas release that will also have to be captured, increasing the volume of radioactive waste. Not all the graphite from HTGR can be reused. In general, the graphite in the fuel blocks will suffer significant activation and will not be suitable for recycling. However, the graphite in the graphite reflectors and structures may be decontaminated and reused. For the graphite reflectors and structures, this would involve mechanical or chemical erosion of the surface of the graphite to remove any contamination from contact with the fission products.

Depending on the graphite separation approach employed, the carbon waste stream will be in the form of carbon dioxide (from oxidation) or graphite (from mechanical separation). In this process, there is the potential for contamination with fission products. If the carbon stream suffered significant contamination, it may be classified as HLW or GTCC waste, and require disposal in a repository depending on its genealogy. In this case, the CO_2 must be trapped and solidified. If the fission product contamination is low enough, the carbon waste can be treated as LLW. Therefore, the wastes from the graphite matrix of spent fuel elements have the potential to be classified as LLW when the proper technology is employed to avoid further contamination of the fission products.

3.3 Other Radioactive Waste Streams

Other radioactive waste streams include stainless-steel components, piping, operating, and decontamination and decommissioning waste streams. Operating radioactive waste includes contaminated equipment; solidified liquid waste; material containers; personal protective equipment; resins and filters; waste cleanup resins; and glove box gloves. In general, these radioactive wastes can be treated and disposed of in the same manner as similar wastes from other nuclear reactors. These waste streams are likely to produce some HLW but with a majority of waste being LLW.

4. CONCLUSIONS

In conclusion, the VHTR design concept is one of the six classes of nuclear reactors in the GenIV initiative, and is the result of several decades of research, development, and operation of HTGR technology. There are two types of HTGRs, the prismatic block-type HTGR and the pebble bed-type HTGR. Both types of HTGRs have TRISO fuel kernels at the heart of the fuel design. Despite very different fuel designs, both types of HTGRs are graphite-moderated, gas-cooled, thermal reactors using many of the same materials. As a result, both prismatic block-type and pebble bed-type HTGRs have similar radioactive waste streams, all of which require safe and secure storage, and eventual disposition. This includes the SNF itself, in core graphite structures and other radioactive wastes.

While many gas-cooled, graphite-moderated, reactors have been built and operated, seven prototypic reactors form the basis of the modern day HTGR designs. These seven model HTGR reactors include the Dragon reactor, Peach Bottom Atomic Power Station (PB), the Arbeitsgemeinschaft Versuchsreaktor (AVR) reactor, Fort St. Vrain Generating Station (FSV), the Thorium High-Temperature reactor (THTR), the High-Temperature Engineering Test reactor (HTTR), and the HTR-10 reactor. Four of the seven HTGR plants constructed and operated, used prismatic block-type fuel; the remaining three HTGR plants used pebble bed-type fuel. Apart from the HTTR and HTR-10, these reactors have all been shut down, the fuel has been placed in safe storage, and the reactors have undergone some degree of decommissioning. As such, there is significant experience in the management of both the SNF, and the radioactive wastes associated with operating these HTGRs. Other graphite-moderated, gas-cooled reactors, such as the British MAGNOX and Advanced Gas-Cooled Reactor (AGR), are able to provide pertinent and noteworthy understanding, despite using carbon dioxide as the coolant in these reactors.

The preferred disposition path for HTGR SNF, depends on the availability and waste acceptance criteria for potential radioactive waste repository. Other factors include the availability and compatibility of the irradiated graphite for disposal as LLW, risk management, and overall cost. The treatment options for the SNF and graphite waste streams depend on the level to which the prismatic-block and pebble-bed fuel can be deconstructed. The simplest approach is whole block or whole pebble disposal. There are also increasingly complex options where these successive layers of graphite structures are removed. This increases the disposal options, but also increases the total volume of radioactive waste. If whole block disposal is unacceptable, or material recovery or waste volume reduction is desired, HTGR SNF can be disposed of following the removal of graphite. Various degrees of prior graphite removal can be performed. Chemical processing of the HTGR SNF can be achieved via conventional processing techniques that allow for the recovery of the U, Pu, and Th (if the Th fuel cycle is used).

The carbon streams from most HTGRs will largely be composed of polycrystalline graphite used as the moderator, reflector, and structural material. The irradiated graphite has significant value for recycle and reuse. Not only does this reuse a valuable material but also greatly decreases the volume of waste to be disposed of. In general, there are three options for graphite disposal: (1) direct disposal without any chemical treatment, (2) graphite gasification, and (3) decontamination and recycling.

Other radioactive waste streams include metal, carbon, operating, and decontamination and decommissioning waste streams. In general, these wastes can be treated and disposed of in the same manner as similar wastes from other nuclear reactors.

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