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***Survey of Nuclear Graphite Waste***

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**COVER PHOTO:** Refueling floor at Fort Saint Vrain HTGR, 1972. Bruce McAllister - U.S. National Archives and Records Administration. Public Domain.

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## 2 Glossary and Abbreviations

ADE-5	Russian production reactor
AGR	Advanced Gas-Cooled Reactor (UK)
AHTR	Advanced High-Temperature Reactor (Alternative description of MSR <i>q.v.</i> )
ANDRA	Agence Nationale pour la Gestion des Déchets Radioactifs, France
ASTM	American Society for Testing and Materials
AVR	Arbeitsgemeinschaft Versuchsreaktor (German HTGR Research Reactor)
BEPO	British Experimental Pile Zero Energy (Early research reactor at Harwell, UK)
BGRR	Brookhaven Graphite Research Reactor
BISO	Bi-Structural Isotropic Particle Fuel
BNFL	British Nuclear Fuels Ltd
BWR	Boiling Water Reactor
CANDU	Canada Deuterium Uranium Reactor
CARBOWASTE	An European Commission collaborative project
CEA	Commissariat à l'Énergie Atomique (France)
CIB	Cured in bed (type of HTGR fuel compact cured in a bed of alumina powder)
CIP	Cured in pile (type of HTGR fuel compact cured during irradiation)
CIRUS	Indian research reactor
CP1	Fermi's original graphite pile in Chicago
CPs	Coated Particles
CPST	Center for Physical Science and Technology (Vilnius, Lithuania)
CRP	(IAEA) Coordinated Research Project
DESNZ	Department of Energy Security and Net Zero (UK)
DIDO	Heavy-water moderated graphite reflector research reactor (of which several examples were built in Europe)
DR-3	Danish 'DIDO' pool-type research reactor
DRAGON	International HTGR Prototype Project based at Winfrith, UK
EdF	Electricité de France (also operators of UK AGRs)
EG	Exfoliated Graphite
EPRI	Electric Power Research Institute (USA)
FIMA	Fissions per Initial Metal Atom
FSV	Fort St Vrain
GDF	Geological Disposal Facility
GIC	Graphite Intercalation Compounds
GLEEP	Graphite Low-Energy Experimental Pile (Early research reactor at Harwell, UK)
GRAPA	'Irradiated <u>G</u> raphite <u>P</u> rocessing <u>A</u> pproaches' – an IAEA collaborative project
GTCC	Greater than Class C Waste (U.S. Category)
HKG	Hochtemperatur Kernkraftwerk GmbH (High Temperature Nuclear Company, Germany)

HLW	High Level Waste
HMC	Heavy Metal Concentration
HOCI	Hydrogen oxychloride
HTGR	High Temperature Gas-Cooled Reactor
HTR-10	High Temperature Reactor, 10 MW (China)
HTR-PM	High Temperature Reactor, Pebble-Bed Module (China)
HTTR	High Temperature Test Reactor (Japan)
HVPF	High Voltage Pulse Fragmentation
IAEA	International Atomic Energy Agency
ILW	Intermediate Level Waste (European Category)
INIS	International Nuclear Information System (IAEA)
INL	Idaho National Laboratory
INPL	Institute of Physics, Université Claude Bernard Lyon 1, France
KONRAD	‘Schacht Konrad’, a former iron-ore mine intended as a repository for intermediate and low-level radioactive waste in Germany
LILW	Low and Intermediate Level Waste
LLW	Low Level Waste
MHTGR-SC	Modular high-temperature gas-cooled reactor – steam cycle
MIT	Massachusetts Institute of Technology
MSR	Molten Salt Reactor
NGNP	Next Generation Nuclear Reactor
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PBMR	Pebble-Bed Modular Reactor of South African design
PGA	Pile Grade A (used in UK Magnox reactor moderator)
PGB	Pile Grade B (used in UK Magnox reactor reflectors)
PIRT	Phenomenon Identification and Ranking Table
PWR	Pressurized Water Reactor
RBMK	Reaktor Bolshoy Moshchnosti Kanalny (High-Power Channel Reactor’)
SiC	Silicon Carbide
SFR	Sodium Fast Reactor
SMR	Small Modular Reactor
SNF	Spent Nuclear Fuel
SoGIN	Società Gestione Impianti Nucleari, Italy
TECDOC	IAEA Technical Document
THTR	Thorium Test Reactor (Germany) <i>also</i> THTR-300 (MW)
TRISO	Tri-Structural Isotropic Particle Fuel
USDOE	United States Department of Energy
VATESI	State Nuclear Power Safety Authority of Lithuania
Xe-100	X-Energy 100 MW HTGR concept

### 3 Executive Summary

This report presents a current state-of-knowledge review of radioactive carbonaceous waste from the operation and decommissioning of graphite-moderated reactors, including high-temperature gas-cooled reactors (HTGRs) and molten salt reactors (MSRs). The scope of the work directly supports the Nuclear Regulatory Commission (NRC) regulatory planning by identifying key safety and technical considerations for carbonaceous waste management, with emphasis on the implications of waste classification (high- vs. low-level), transportation, and storage. The report underscores that not all irradiated carbonaceous material is alike, and its physical form, contamination level, and neutron exposure history significantly affect how it should be managed.

The report is divided into a number of sections, following the executive summary: 4) Introduction, 5) Information from Legacy Graphite, 6) Nuclear Graphite and its Irradiation Behavior, 7) Planning the Decommissioning and Disposal of Carbonaceous Waste, 8) Potential Treatments for Graphite Waste, 9), Fuel Matrix Material, 10) Separation of Spent TRISO Fuel from Fuel Elements (covering various technologies), 11) Summary.

Irradiated graphite, graphitic matrix<sup>1</sup> and baked carbon undergoes complex changes over time, including dimensional distortion, cracking, accumulation of activation products (notably <sup>14</sup>C and <sup>36</sup>Cl), and entrapment of fission products and metallic contaminants from the reactor coolant circuit. Structural integrity has degraded significantly in legacy reactors. The presence of radioactive dust is especially problematic in pebble-bed reactor designs, and the dust often carries high concentrations of fission products (e.g. <sup>137</sup>Cs, <sup>90</sup>Sr), activation products and actinides.

The behavior of irradiated graphite is highly dependent on reactor design. Pebble-bed HTGRs generate more contaminated dust and fuel element debris than prismatic designs. MSRs that incorporate graphite moderator blocks or matrix “graphite” in the fuel may face additional chemical and contamination challenges from molten salt interaction. MSRs using FLiBe or FLiNaK salts introduce new challenges to graphite management due to potential salt contamination of the graphite, raising questions about criticality control, corrosion products, and chemical stability during transport and storage. This report highlights that future reactors are likely to produce more operational graphite waste (from fuel elements) than end-of-life structural graphite (reflectors and moderator stacks).

The classification of graphite waste - as low-level waste (LLW), greater-than-class-C (GTCC), or high-level waste (HLW) - has implications for transport, interim storage, and disposal. The report notes that much of the graphite waste, particularly from spent TRISO (Tri-Structural Isotropic) fuel elements and fuel-adjacent graphite, will likely exceed LLW thresholds due to long-lived beta emitters such as <sup>14</sup>C. Safety measures will likely include robust shielding, containment, and possibly geological storage or disposal.

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<sup>1</sup>The term ‘Graphitic Matrix’ is used in this report when specifically addressing the carbonaceous material which make up the fuel elements and contains the TRISO particles. Spherical fuel elements (pebbles) or cylindrical fuel rods (compacts) inserted into graphite blocks are considered to be fuel elements.

Efforts to reduce activation may help lower waste classification but are limited by fundamental physics (e.g., production of  $^{14}\text{C}$  by route  $^{13}\text{C}(n, \gamma)^{14}\text{C}$  which is unavoidable with  $\sim 1.1\%$   $^{13}\text{C}$  in all-natural carbon material). Strategies being investigated include direct whole-block disposal as well as volume-reduction techniques (such as oxidation, chemical disintegration, or mechanical fragmentation) to enable more cost-effective waste categorization.

This report examines graphite waste, considering dismantling, characterization, treatment, and final disposal as an integrated lifecycle.

This state-of-knowledge report provides a foundational understanding of the complexity, variability, and potential risks associated with irradiated graphite waste. The key considerations are the variety of carbonaceous waste streams due to functional roles and reactor types, the benefit of early-stage integration of waste handling and disposal planning into reactor design licensing, and the importance of graphite and matrix graphite characterization for determining appropriate waste classification and treatment methods.

This report helps prepare the NRC staff for efficient licensing reviews of graphite waste storage and transportation for the increased and differing carbonaceous wastes expected from advanced reactors.

## 4 Introduction

The objective of this report is to gather information on nuclear graphite irradiation behavior and radioactive carbonaceous waste management, to enable the U.S. Nuclear Regulatory Commission (NRC) to have a comprehensive understanding of the issues that will become relevant when managing the graphite and graphitic matrix waste streams from proposed high-temperature reactor (HTGR) or molten salt reactor (MSR) designs, including some designs which fall under small modular reactors (SMRs).

This report's effort is inclusive of experience with fuel elements containing Tri-Structural Isotropic (TRISO) fuel particles, which themselves comprise approximately 70-90% carbonaceous material (Vrinda Devi *et al.* 2019). There is significant industry interest in using TRISO particle-based fuel designs (Demkowicz 2024). In most designs using TRISO fuel particles, the TRISO fuel particles are incorporated into larger structures comprised of carbonaceous graphitic matrix which are manufactured from natural graphite mixed with organic resinous material and then baked.

In this report, the terms 'nuclear graphite' and 'graphite' generally refer to graphite that was prepared industrially for use in nuclear reactors. It is useful to make the point that 'nuclear graphite', whilst carefully manufactured and purified to high standards, is not 'pure' graphite in the chemical and physical sense (the allotropic structure with parallel layers of carbon-atom hexagons) but is an industrial material produced from pitch and coke which is extensively, but incompletely, graphitized: the micro-crystallite structures are distorted and there are regions of 'mesophase.' Nuclear graphite generally has a purity level better than five parts per million boron equivalent and a density greater than  $1.50 \text{ g/cm}^3$  (IAEA 2002). Figure 1 shows this complex structure at increasing magnifications from a reactor component down to the individual layered crystal structure. The material is also porous, with both closed and open porosity (the latter accessible to the surrounding gas atmosphere) which can amount to 20-25% of the total volume. A typical density of nuclear graphite is  $1.7 - 1.8 \text{ g/cm}^3$  compared with the crystal density of  $2.26 \text{ g/cm}^3$ . Henceforth, we use the term 'graphite' to mean this industrial material.

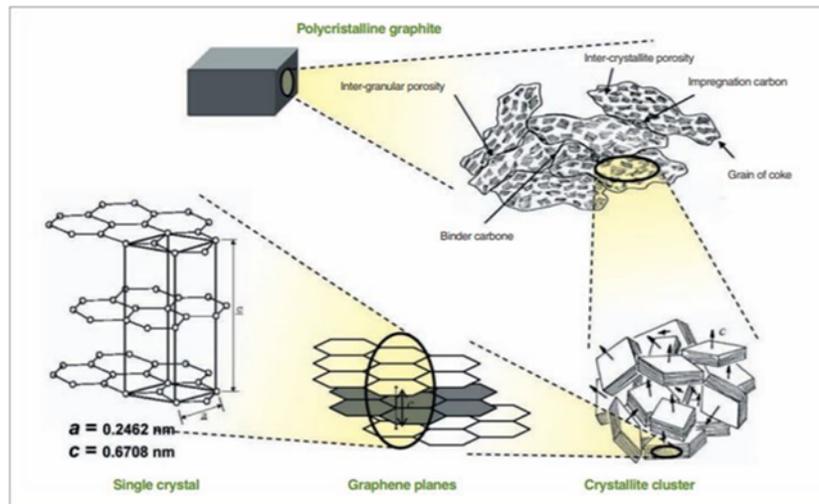


Fig 1: Lattice structure of industrial graphite at increasing magnifications (from Rouzaud *et al.*, 2012)

As noted above, designs using TRISO fuel particles generally involve the particles being incorporated into larger structures, with the larger structures typically composed of nuclear graphitic

matrix. The temperatures these larger structures are heat-treated to are high (around ~2000 C) but are not as high as to achieve maximum graphitization of the resin, as doing so would damage the structures which encompass the TRISO particles. Two common design choices are to incorporate the TRISO particles into ‘pebbles’ or ‘cylindrical compacts in hexagonal prismatic graphite blocks’, where the pebbles are considered fuel elements and the prismatic blocks are also considered fuel elements (Torres et al. 2025).

Torres et al. note that: “TRISO is currently the coated particle fuel form of choice for next generation HTGRs and very high-temperature reactors. This former designation includes pebble bed reactors and prismatic core reactors, which are differentiated by the forms encasing the TRISO particle fuel. Pebble bed reactors utilize spheres that are typically approximately 6 cm in diameter. Prismatic core reactors utilize cylindrical-type fuel compacts designed to fit into hexagonal graphite blocks, which are referred to as prismatic blocks. In the United States, the design compacts for prismatic blocks are generally 12 mm in diameter and 49 mm long. By convention, both pebbles and prismatic blocks are discrete units that are referred to as fuel elements, while the cylindrical compacts are not referred to as fuel elements. The pebbles are not immobilized in any larger structure and thus differ from the hierarchical fuel element structure in today’s LWRs. In contrast, the cylindrical compacts and hexagonal block fuel elements have a hierarchical structure, analogous to how today’s LWR fuels consist of multiple rods contained in discrete assemblies.” Thus, the fuel elements in TRISO reactors will involve both nuclear graphite and graphitic matrix that are subject to irradiation, for all designs currently expected.

Information on graphite irradiation behavior comes principally from the legacy graphite (UK Magnox and Advanced Gas-Cooled Reactors (AGR) and Russian-designed RBMKs<sup>2</sup> which are essentially boiling-water reactors moderated principally by graphite). This legacy-graphite experience provides valuable background knowledge relating primarily to the moderator and reflector graphite in new designs and represents a ‘one-off’ waste stream which is only important at the end of reactor life. The fuel element stream, however, is ‘operational waste’ which will be produced continuously during the period that the reactor is at power and has the potential to become a much larger volume of material to be managed (Forsberg 2024, Kim 2024).

Figure 2 provides the ‘logic’ of irradiated graphite waste management, recognizing that the diagram may require modification for specific scenarios. As discussed in more detail later in this report, interim storage for irradiated graphite and graphitic matrix components (such as irradiated fuel blocks or pebbles, moderators and reflector blocks), may not be the ideal route. Particularly when one considers options for potential treatments (discussed below) which could beneficially impact waste categorization or volume. Additionally, interim storage strategies may become less appealing if final disposal facilities become already available. For the operational waste, interim storage as the first stage is probably unavoidable. The categories in Figure 2 are based on IAEA classification, and those categories do not directly align with the waste classification in the U.S., though the same general principles and flow of the figure would apply in the U.S. waste classification system.

The diagram exhibits both waste streams and their inter-correlation when separating irradiated graphite or graphitic matrix from the fuel element structure. In the U.S. waste classification system, this could be foreseen for reducing the waste volume of the spent fuel and for isolating HLW of the

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<sup>2</sup> Reaktor Bolshoy Moshchnosti Kanalny (‘High-Power Channel Reactor’) of Soviet design, originally deployed in the Russian Federation, Ukraine, and Lithuania

spent fuel from the structural carbon material, which may fall into lower waste categories. Even though conditioning, packaging and disposal of HLW from spent fuel is not the subject of this report, technical options for the separation of fuel compacts from the graphite fuel element blocks and from TRISO particles extracted from the spherical fuel element matrix are mentioned (yellow boxes) as they do influence waste management of irradiated graphite waste and graphitic matrix waste.

Sorting the carbonaceous waste streams is an important procedure because the radiological activity of the related waste streams will be different according to the irradiation history and environments. Laboratory scale treatments of irradiated graphite have shown that most radioactive contaminants can be reduced. This also includes  $^{14}\text{C}$ , which is preferably deposited at the surface of the carbon components and in the pore structure. These purification or decontamination treatments (included among the grey boxes) may be necessary for the  $\text{CO}_2$  storage route and for a classification of irradiated carbon material into lower waste categories. However, only a few such exploratory treatment tests have been or may be conducted to date, using irradiated graphite stemming from operated HTGRs, as there is no facile access to representative sample material for testing and advancing such treatments.

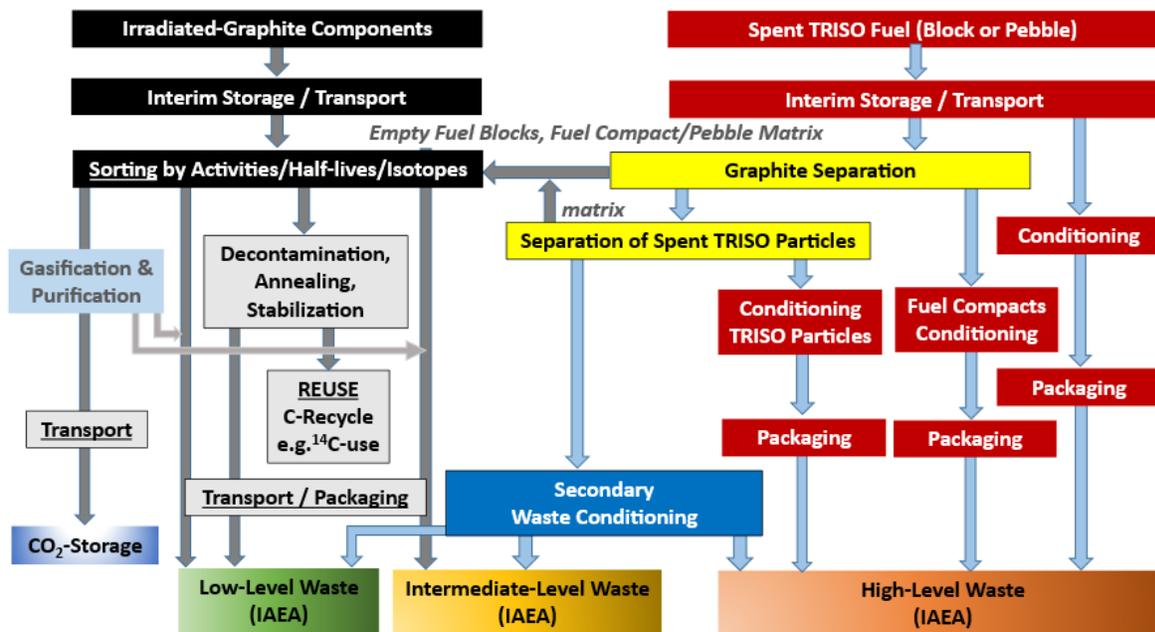


Figure 2: Provisional logic diagram addressing the options for waste streams of moderator/reflector material (black starting point) and operational waste (red starting point) from fuel containing TRISO Fuel particles

It is worth mentioning that high-temperature treatments also support the annealing of irradiation defects. This will also lead to a stabilization of the radioactive isotopes within the restored graphite structure. Graphite itself is very stable and more corrosion-resistant than most metals in the long-term.

## 5 Information From Legacy Graphite

Since the first days of the investigation of criticality in Fermi's reactor stack 'CP1' in Chicago in 1942, graphite has been viewed as a potential material for moderating neutron energy to enable a

fission chain reaction to occur in nuclear fuel. Reactors such as those at the Hanford Site in the USA, at the Windscale Site in the UK, as well as others in Russia, China and in France, were initially designed for the production of weapons-grade plutonium, before designs morphed into large power-production reactors such as the Magnox and AGR in the UK, the Soviet RBMK design, and the UNGG<sup>3</sup> reactors in France.

The principal alternative moderator for thermal power reactors is water (or, in a minority of cases, heavy water, D<sub>2</sub>O), with boiling-water reactor (BWR) and pressurized-water reactor (PWR) designs employed widely around the world, plus the CANDU (Canada Deuterium Uranium Reactor) heavy-water design found in Canada, some Asian countries, and Romania. The aforementioned RBMK design is also a BWR, with its fuel immersed in water tubes passing through the graphite stack, and so is, in fact, ‘dual-moderated’.

Currently, graphite is being employed as a neutron reflector and moderator in the high-temperature reactor ‘pebble-bed module’ (HTR-PM) in China and is being considered for other HTGR designs in the current roll-out of modular HTGR reactor concepts internationally, together with MSRs (IAEA, 2022). Two HTGR test reactors are operational (High Temperature Test Reactor [HTTR] in Japan, HTR-10 in China).

The advantages of employing graphite were, and are, at least four-fold:

- It has favorable neutron-moderation<sup>4</sup> characteristics and the capability to reflect neutrons back from the peripheral core regions
- It is a strong solid material and therefore can form the basis of the reactor structure with high load-bearing capability.
- It has a high heat capacity, which is important in safety-case assessments for emergency situations.
- It is physically and chemically stable to high temperature and can withstand the rigor of such an environment for long-term irradiation.

However, graphite and graphitic matrix are subject to irradiation damage that results in diverse property changes. The material behavior related to irradiation must be understood and managed:

- Fast neutrons interact with the carbon atoms in the crystallite structure in the course of moderation, leading to displacements and crystalline rearrangements on the lattice scale.
- Atomic-scale changes also occur involving alterations in the arrangement and displacement of individual atoms. They both lead in turn to changes in the physical and mechanical properties of the graphite as irradiation proceeds, such as dimensional change, systematic changes in strength, Young’s modulus, thermal conductivity and the coefficient of expansion.
- Figure 3 illustrates the systematic distortion of layers and production of interstitial bonding, which results during low temperature irradiation of graphite (highest damage rate). The units are in displacements per atom (dpa). Commercial power reactors have accumulated well in

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<sup>3</sup> Uranium Naturel Graphite Gaz, comparable with UK Magnox designs but with different configurations

<sup>4</sup> Moderation is effectively energy reduction, enabling the fission chain process to propagate.

excess of 20 dpa, albeit at higher temperature irradiation, and so it will be understood that the crystallites become severely ‘altered’ during irradiation. It may be considered remarkable that the irradiated components retain their original geometry and strength (within the limitations already mentioned) until eventually (as in UK AGRs), differential stresses lead to cracking.

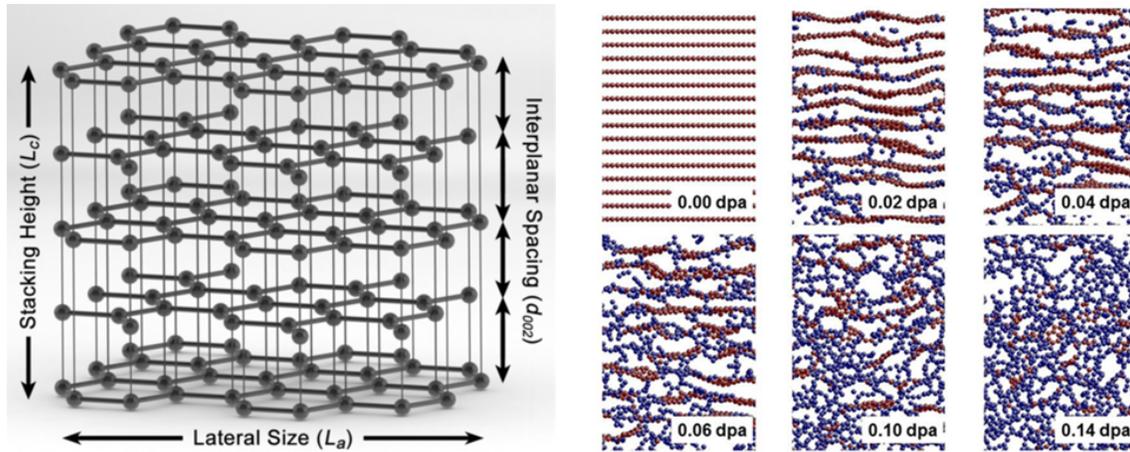


Fig 3: Illustration of the effect of fast-neutron irradiation on the initially lamellar structure of a graphite crystallite at low irradiation temperature (from Chartier *et al.* 2018)

- Greater detail on these structural change issues can be found in Rouzaud *et al.* (2015), and the interested reader is referred to the extensive computational chemistry work on bonding and layer changes under irradiation, such as ‘buckle, ruck and tuck’ – see, for example Heggie *et al.* (2011).
- The ‘thermalization length’ (Thermal Neutron Diffusion Length) for fast neutrons in graphite is quite large with about 50.95+/- 0.5 cm in comparison to light or heavy-water (Mirfayzi 2013). This results in much larger dimensions and volumes/mass for the reflector and moderator structures.
- In early reactors, where irradiation has proceeded at temperatures generally below about 200°C, accumulation of stored energy occurred though the introduction of crystal defects (‘Wigner energy’) and which can be managed (in the worst cases) to avoid inadvertent energy release: this is not an issue for reactors now under consideration (Gallego and Burchell 2011).
- A particular issue with fast-neutron damage is the development of differential stresses within components, dependent upon their distance from fuel elements (flux gradient) and which, although alleviated to some extent by irradiation creep, can potentially induce cracking initiated at stress-raising features and which may therefore determine the operational life of the reactor stack, as in the UK AGRs. Additionally, they may lead to distortion of fuel and control-rod channels (‘bowing’) and in some designs (such as RBMK) to whole-core displacement and potential jamming of individual graphite components when dismantling is to be undertaken.
- At temperatures relevant to future reactors, graphite first shrinks under irradiation because microcracks arising in the graphitization process and some of the porosity (known as

“accommodation porosity”) is available to accommodate expansion in the crystallographic ‘c’ direction (orthogonal axis) alongside the shrinkage in the ‘a’ direction (basal plane); simultaneously, some existing pores close up whilst new ones are created, this all largely controlled by local stresses developing in the material. The rate of shrinkage decreases with continuing irradiation, and a point known as ‘turn-around’ is reached, beyond which the graphite expands again. If sufficient continued irradiation is possible, commercial reactors may reach a point at which the gaps between graphite components approach zero. Some reactor designs cannot operate beyond the point where the dimensions return to the original values: this arises for RBMK reactors were built with no gaps between columns and thus stresses would quickly lead to component failure in those reactors. Now that the dimensional change behavior of nuclear graphite is much better understood, stacks for modern designs can be developed which will accommodate any anticipated expansion.

- Slow neutrons (mainly, but not exclusively) activate certain impurity atoms in the graphite to create radioisotopes. Direct radioactive contamination may occur from this activation. Indirect radioactive contamination may arise from particulate materials from other parts of the reactor circuit (e.g. heat exchanger corrosion), which may become trapped in the graphite pores and then become activated. In addition, long-lived beta-emitting  $^{14}\text{C}$  from the 1.1%  $^{13}\text{C}$  present in the material arises through an  $(n, \gamma)$  reaction together with additional  $^{14}\text{C}$  production from any nitrogenous ( $^{14}\text{N}$ ) residues remaining from the pitch (mainly quinolines) and any entrained nitrogen (in closed pores) from the graphitization process ( $n, p$  reaction). Other weak beta-emitting isotopes likely to be associated with the graphite are  $^{36}\text{Cl}$  and tritium.
- Where exposed to an oxidizing coolant, graphite can partially oxidize to carbon dioxide through a radiation-induced or a chemical process, resulting in progressive reduction in density (“weight loss”). For example, graphite may be oxidized in presence of carbon dioxide to carbon monoxide by the Boudouard reaction ( $\text{C} + \text{CO}_2 \rightleftharpoons 2\text{CO}$ ). Carbonaceous deposits can also be produced on graphite surfaces as the concentration of the oxidation product, carbon monoxide, is allowed to rise in the coolant, *via* the reverse Boudouard reaction ( $2\text{CO} \rightleftharpoons \text{C} + \text{CO}_2$ )<sup>5</sup>. In pebble-fuel designs, the major source introducing oxidizing impurities is the pebbles themselves, introducing adsorbed water which also reacts with graphite under operational conditions. For the Magnox reactors, the temperature and conditions are chosen to keep the forward Boudouard reaction rate low.
- Radiolytic corrosion of carbonaceous material under storage conditions also needs to be taken into account in presence of strong ionizing radiation fields and oxygen, humidity, or both.

Information regarding the structural integrity of the graphite in different legacy reactor systems at the end of their operational lifetimes is potentially of interest in evaluating modern designs; because the component lifetime, as well as the removal and packing of the waste, are affected by the structural integrity. There are no known structural failures in western production reactor stacks nor in UK Magnox reactors (which employed Pile Grade A [PGA] graphite as moderator and Pile Grade B [PGB] as reflector) with the exception of the very bottom layers of graphite in Calder Hall and

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<sup>5</sup> In line with current trends in carbon capture and energy storage, this reaction is being investigated in that role using plasma processing powered by renewable energy (Li *et al.* 2020)

Chapelcross where some distortions are evident from low temperature dimensional change and a greatly-extended operational lifetime – there are no known failures in the subsequent commercial Magnox reactors, again despite operation significantly beyond the design lives.

In contrast, UK AGRs (using Gilsocarbon graphite) have suffered extensive component cracking across a number of stack layers, generally as whole-length vertical cracking arising at stress-raising features in the machined components, with some limited fragmentation which is a potential concern for fuel-stringer movement. Again, these reactors have operated safely well beyond their original design lifetimes, and eight remain in service with regulatory approval which recognizes the resilience of the keyed structures to seismic events even when with a high population of single-cracked bricks and a smaller population of double cracks (Reed 2023). It is worth noting that limited areas of AGR graphite stacks have experienced in excess of 40% mass loss through radiolytic oxidation in carbon dioxide.

RBMK reactors have consistently experienced significant channel distortions due to ‘barreling’ of individual components in the higher-flux regions introducing lateral forces upon the core as a whole (Leshchenko 2023).

Figure 4 indicates the location and estimated quantities of the irradiated graphite legacy world-wide. The majority of this graphite resides currently in undisturbed reactor stacks within their original containments: only a few reactors have been dismantled. In most of those cases, the graphite has gone into ‘temporary storage’, often in a newly constructed building, because the final destiny of the material has not been decided. Another option which mitigates radiation exposures has been leaving the graphite in the reactor containment until final disposal and recycling/reuse pathways are available.

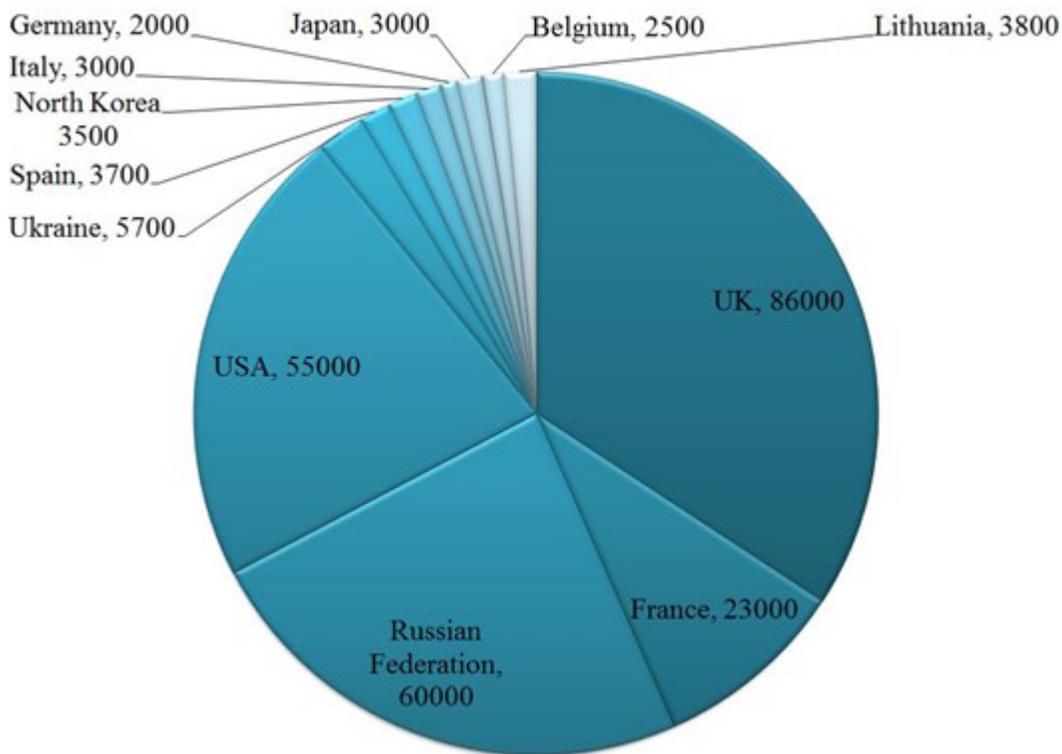


Figure 4: Location of Legacy Irradiated Graphite World-Wide. Tonnages are estimates and subject to upward revision. Some minor sources are omitted for clarity. (from Wickham *et al.*, 2017)

In addition to reactor stacks themselves, there is a significant amount of graphite debris from other sources – for example, fuel element components and some control-rod assemblies – and in some cases these wastes have been placed in on-site vaults with other material<sup>6</sup>, which adds considerably to the difficulty if these materials need to be segregated again for final disposal.

Any future roll-out of advanced reactors would likely generate considerable quantities of irradiated graphite and carbonaceous materials: mostly reflector graphite and spent fuel matrix from HTR types. Graphite moderators from MSR designs may also be contaminated with the salts (‘FLiBe’ or ‘FLiNaK’) and, from those designs where the fuel is present in the molten salt itself, actinides and their fission products etc.

Proposed SMR designs have either secondary circuits to separate the fuel from the graphite or may use fuel similar to HTGR. A valuable review of the overall waste issues associated with future small reactors can be found in Krall *et al.* (2022) although, in terms of graphite, it relies largely upon an earlier review by Fuks *et al.* (2020). This 2020 review comes from the National Center for Nuclear Research in Poland and makes reference to the extensive International Nuclear Information System (INIS) database of the International Atomic Energy Agency (IAEA). Broadly, the Fuks review makes similar points about the quantities and perceived management difficulties for irradiated graphite and

<sup>6</sup> A typical example, from the UK, has seen graphite ‘splitters’ from fuel elements admixed with Magnox and other potentially radioactive metal in vaults into which water ingress has subsequently occurred. This was done, apparently, without planning eventual disposal.

notes the potential contribution which would arise from the spent fuel pebbles of HTGRs. The principal conclusions of the Krall review reinforce the importance of directly considering the waste-management of advanced reactors:

*“This analysis of three distinct SMR designs shows that, relative to a gigawatt-scale PWR [Pressurized Water Reactor], these reactors will increase the energy equivalent volumes of spent nuclear fuel [SNF], long-lived (low and intermediate-level waste, LILW), and short-lived LILW by factors of up to 5.5, 30, and 35, respectively. These findings stand in contrast to the waste reduction benefits that advocates have claimed for advanced nuclear technologies.”*

The quotation was based on an analysis that considered the entirety of the potential waste streams, and not just graphite and carbonaceous materials; the operational waste (spent fuel pebbles and blocks) forms the major stream.

All of the irradiated graphite and carbon from the legacy reactors and those of the future will require carefully considered management. Initiatives associated with SMR development include careful appraisal of the graphite manufacturing process to improve the chances of the resulting waste being classified as LLW.

It is important that the lessons learned from consideration of legacy graphite are understood when considering future reactors. This report also seeks to outline the current state-of-knowledge in the areas of characterization, removal/retrieval, potential treatments and (to a limited extent) packaging and disposal, in order to assist the planning of a disposal process. Research on these topics has involved a large number of specialist researchers internationally and forms the basis of the most recent IAEA Technical Document (‘TECDOC’) on the subject (IAEA, 2024): this reference reports the results of a four-year international collaboration “Irradiated **Graphite Processing Approaches**” (‘GRAPA’; Wickham *et al.*, 2017) and is the latest in a succession of IAEA publications detailing similar Collaborative Research Programs (CRPs) and conferences featuring work in this area (IAEA 2001, 2006, 2010, 2016). Note that these publications only cover topics which have been offered by the various Member States and are therefore not necessarily comprehensive. Other useful reference publications include a sequence of reports prepared for The Electric Power Research Institute (EPRI) (Wickham and Bradbury 2006, 2007, 2008, 2010, 2012, Bradbury and Mason, 2008, Bradbury and Goodwin 2010) and the EU CARBOWASTE project, a European Commission collaborative project, (Grambow *et al.* 2013 and Wareing *et al.* 2013, 2017).

Figure 5 provides a graphic timeline for most of the international activity, whilst the EPRI and other U.S. initiatives (not shown) took place between 2006 and 2012, in parallel with and after the EU CARBOWASTE project.

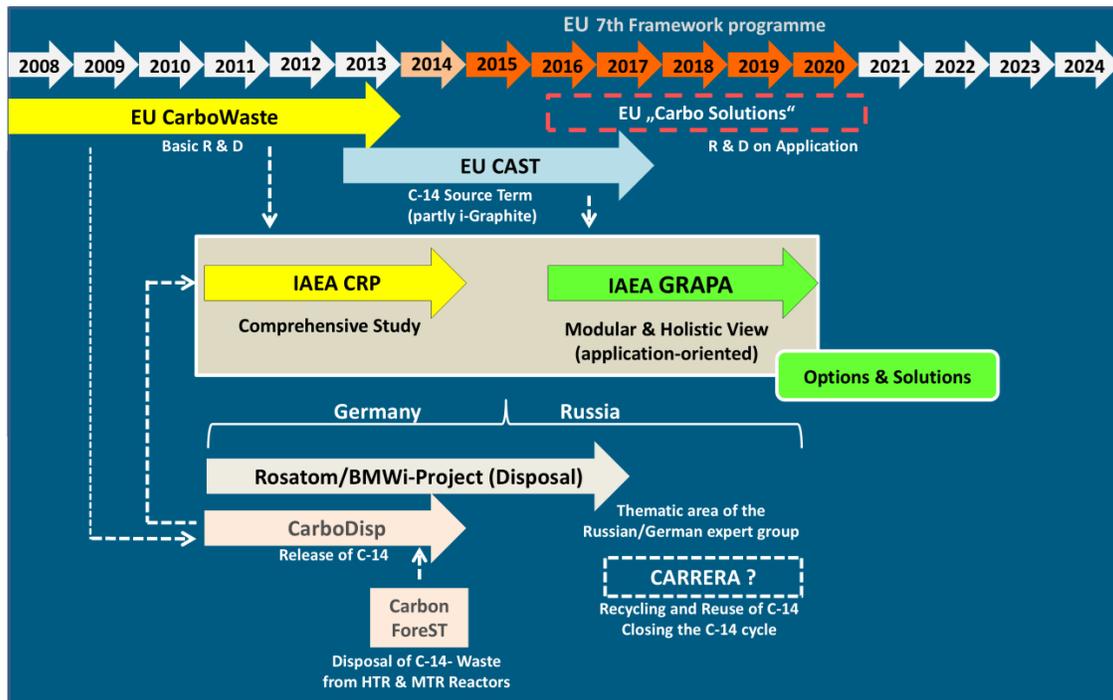


Figure 5: International Initiatives to Address the Management of Irradiated Graphite (IAEA, 2024).

The first consideration of the management of irradiated graphite waste appears to have been over 40 years ago in a study conducted for the Commission of the European Communities (forerunner of The European Union) (White *et al.* 1984): the progressive closure of graphite-moderated reactors since that time has brought about a renewed focus on international initiatives, which includes alternatives to deep geological disposal.

Additional research activities on ‘Reduction, Mitigation and Disposal Strategies for the Graphite Waste of High Temperature Reactors’ have been launched in the U.S. including a related roadmap (Forsberg 2024).

## 6 Nuclear Graphite and its Irradiation Behavior

### 6.1 Graphite Production

No two sources of irradiated graphite are ever the same and there is no ‘one size fits all’ solution to their waste management strategies. The specifications of nuclear graphites have evolved with time and, for the next generation of reactors, such graphites tend to be ‘designed’ to maximize their performance and stability in the evolving reactor environments. Knowing the provenance of the graphite, and the conditions under which it has been exposed during irradiation, are essential in formulating a dismantling and waste management strategy.

Graphite is fundamentally a mix of ground coke (whose source may be pitch, petroleum or directly extracted mineral such as Gilsonite) with pitch and usually some proportion of ‘filler’ which may be derived from graphite from previous productions after the machining of components. This so-called ‘green mix’ is formed into the shapes required either by extrusion (as generally used in the manufacture of graphite electrodes) or molding. Molding techniques for nuclear graphite components have evolved, from simple axial pressing to isostatic molding and thence to vibration molding, all

with the objective of reducing anisotropy. There follows a baking stage to form the basic solid material which may be further impregnated with pitch and re-baked to increase the density. This may happen more than once. The baking is usually carried out around 1200 °C to 1400 °C and involves the volatilization of binders into carbon. The baked article is then graphitized at about 3000 °C, when the carbon atoms are rearranged from disorder into a graphite crystalline order. Graphitization is often conducted by the traditional Acheson process, in which electric currents are passed through the stacked components whilst they are blanketed in packing coke. The coke serves as an oxygen ‘getter’, assists the path of the electric current, and helps to even out the temperature distribution in the furnace. After cooling, the blocks can be machined to the exact specification required for the reactor. For high performance nuclear-grade graphite, a significant amount of purification<sup>7</sup> is required. Originally this was conducted with chlorine gas, and then with freons, to remove neutron absorbing boron and heavy metals as their volatile halides. More recently, magnesium fluoride has been employed since it is a vapor at the graphitization stage (temperatures of order 2700°C), with which purification can then be combined. It has the advantage that neutron-activation of the residues in the finished products is minimal.

Minimizing the impurity content of nuclear graphite is important for three reasons, with each reason corresponding to a different type of impurity. The first is to ensure that the concentrations of elements which have isotopes capable of absorbing neutrons (exhibiting high neutron-capture cross sections), such as boron, are at sufficiently low levels to avoid compromising the nuclear reactivity of the reactor core. This is often expressed as the ‘boron equivalent’ metric for neutron absorbing impurities. The second is to keep to a minimum the concentration of any elements which may become activated to longer-lived isotopes which might then act as significant radiation sources and may compromise future operations (e.g. maintenance inspections) or the eventual dismantling and disposal of the graphite stack. In very early reactors, this latter issue was given great importance by the specialists of the day: later, standards were introduced which have actually led to significant difficulties because the standards permitted concentrations were less restrictive. In some of the UK AGRs, for example, there is a high gamma flux from the graphite (arising primarily from <sup>60</sup>Co) which will take around 50 years (approximately ten half-lives) to decay to safe levels after the end of the operating period. The third reason is to reduce the presence of any fissionable impurities that may be present during graphite manufacturing (such as uranium and thorium, which can be present at levels on the order of 1 ppm in natural graphite): during reactor operation these fissionable impurities and their subsequent fission products and transuranic activation products are partially retained in the graphite structures (and partially released into the primary cooling circuit).

Research on understanding the details of impurities in legacy and currently available graphites has been greatly assisted by advances in analytical techniques such as electrothermal vaporization, prompt-gamma neutron-activation analysis, inductively coupled plasma mass spectrometry and glow-discharge mass spectrometry. Of the impurities mentioned in the previous paragraph, uranium, thorium and nitrogen are often not measured and quantified (see Table 1).

Studies have been performed on different grades of nuclear graphite. Grades of nuclear graphite are not defined by standards developing organizations and are simply manufacturer labels that are based on composition, performance characteristics, and intended applications. As these are manufacturer

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<sup>7</sup> Some manufacturers delay purification until after the primary graphitisation. High temperature would need to be maintained in order to create the volatile halides from most relevant impurity elements. At least one manufacturer prominent in the potential supply chain for USA SMRs continues to use chlorine.

determined labels, such that a manufacturer could make changes to a composition while continuing to use the same 'grade' on the product. The results from studies can be interpreted by recognizing that they were performed on the 'grade' used at that time. Table 1 shows the results of a recent comparison of graphites from a range of legacy sources alongside current production material being assessed for use in SMRs, utilizing glow-discharge mass spectrometry. The column headings (other than Sample name) refer to grades of nuclear graphite.

Table 1: Comparison of Impurity Concentrations (units of ppm) in a Range of Graphite Sources: ET-10, ETU-10 and NBG-18 are currently under consideration for utilization in U.S. SMRs (from Rani et al., 2024). The column headings (other than Sample name) refer to grades of graphite (see text for additional explanation).

Sample name	ET-10	ETU-10	CP-1	H-451	IG-110	IG-11	NBG-18	NBG-25	PCEA	PCIB	HANFORD	PGA	WINDSCALE	RBMK
Element	Conc.	Conc.	Conc.	Conc.										
Li	< 0.01	< 0.01	0.05	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01	< 0.01	0.05	< 0.01	< 0.01	< 0.01
B	0.62	0.34	0.5	1.3	0.04	1.6	5.7	0.01	1	0.86	0.38	0.09	0.11	0.32
Na	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.05	< 0.05	< 0.05	< 0.05	0.34	< 0.05	2.6	< 0.05
Mg	< 0.05	< 0.05	0.05	< 0.05	< 0.05	0.06	0.05	< 0.05	< 0.05	< 0.05	1.8	< 0.05	2.9	< 0.05
Al	0.06	< 0.05	0.23	< 0.05	< 0.05	< 0.05	1.6	< 0.05	3.2	< 0.05	0.08	< 0.05	0.96	< 0.05
Si	1.3	1	21	5.9	2.7	0.42	3.9	0.86	8.7	17	9.9	6.5	15	1.2
P	1.4	0.72	0.98	0.35	0.34	0.12	0.54	0.13	0.15	0.17	0.4	0.33	0.8	0.3
S	80*	8*	80*	20*	5*	30*	200*	4*	30*	4*	130*	21*	80*	7.4*
Cl	1.4	1.1	2.6	0.89	0.51	1.2	0.3	0.42	0.24	0.4	28	1.8	1	1.4
K	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.82	0.6	0.68	0.59
Ca	2	< 0.05	12	< 0.05	< 0.05	0.44	5.7	< 0.05	3.3	< 0.05	14	0.65	2.9	< 0.05
Sc	0.01	< 0.01	0.01	< 0.01	< 0.01	0.01	0.01	< 0.01	0.02	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Ti	0.6	< 0.01	1.2	0.02	< 0.01	0.41	0.97	< 0.01	1.3	< 0.01	0.94	0.14	0.26	0.02
V	0.08	< 0.01	19	0.03	0.01	8.4	0.1	< 0.01	29	0.01	1.1	0.41	0.34	< 0.01
Cr	< 0.1	< 0.1	0.33	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.14	< 0.1	0.56	0.15	< 0.1	< 0.1
Mn	< 0.01	< 0.01	0.01	0.01	< 0.01	0.01	0.01	< 0.01	0.02	< 0.01	0.02	0.02	0.15	< 0.01
Fe	1	0.01	3	0.09	0.02	0.26	0.72	< 0.01	41	0.01	0.28	1.3	2.1	0.13
Co	0.01	< 0.01	0.01	0.01	< 0.01	0.01	0.02	< 0.01	0.44	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Ni	0.15	< 0.01	1.1	0.06	< 0.01	0.98	0.09	< 0.01	23	< 0.01	0.05	0.13	0.11	< 0.01
Cu	< 0.05	< 0.05	0.06	0.06	< 0.05	0.06	< 0.05	< 0.05	< 0.05	0.06	< 0.05	0.09	0.32	< 0.05
Zn	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.14	0.09	< 0.05	0.07	0.07	0.13	0.06	5.1	< 0.05
As	0.07	< 0.05	0.11	< 0.05	< 0.05	< 0.05	0.15	< 0.05	< 0.05	< 0.05	0.23	0.12	< 0.05	< 0.05
Zr	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.08	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
Mo	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.16	< 0.05	< 0.05	0.44	< 0.05	0.16	< 0.05	< 0.05	< 0.05
Cd	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.19	< 0.05
La	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.06	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Ce	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	< 0.01	< 0.01	0.03	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
W	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.07	< 0.05	< 0.05
Pb	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	0.39	< 0.05

There have been previous similar intercomparisons – for example, Berlioux and Barth (2008) in a round-robin study for ASTM (American Society for the Testing of Materials).<sup>8</sup> IAEA (2024) *Annex 1* contains a detailed study on British graphites conducted by The University of Manchester, together with a wealth of historic impurity data, but it should be borne in mind that the analytical techniques used in early work were less sensitive and less accurate than modern methods.

A considerable effort has been put into understanding the required mechanical and physical properties of graphites for satisfactory performance in future reactors. Three papers from the 23<sup>rd</sup> International Nuclear Graphite Specialists Meeting illustrate this point: Arregui-Mena *et al.* (2023) made extensive comparisons of the microstructural features and mechanical/physical properties of currently available candidate graphites; Campbell *et al.* (2023) examined graphite irradiation behavior in the specific context of the X-Energy 100MW HTGR concept (Xe-100 HTGR); Gallego *et al.* (2023) offered a similar analysis of the materials challenges for graphites utilized in the molten salt environment.

<sup>8</sup> This was mentioned in a USNRC report under NUMARK Associates contract US-HQ-25-14-E-0004, August 2021 – ‘Appendices to the Assessment of Graphite Properties and Degradation, including Source Dependence’ (Srinivasan *et al.* 2021)

## 6.2 Activation During Reactor Operation

The isotopes of concern in irradiated carbonaceous material can broadly be divided into three groups:

- isotopes presenting a safety issue in accident scenarios during reactor operation;
- those which present an immediate problem during reactor maintenance or dismantling, but have relatively short half-lives (these are principally gamma emitters); *and*
- those with longer half-lives which present issues in temporary storage, the so-called ‘operational phase’ of long-term repositories (when transferring to a geological disposal facility for example) and perhaps also in the event of a breach of containment in the future. These are primarily weak beta emitters.

The formation of  $^{14}\text{C}$  from the  $^{13}\text{C}$  in natural carbonaceous materials is unavoidable despite the relatively low capture cross section. Efforts are currently being made by graphite manufacturers in the USA to produce nuclear graphite for the next generation reactors which will develop a lower  $^{14}\text{C}$  content than previously by lowering the nitrogen content as much as possible (to avoid the production of  $^{14}\text{C}$  via the  $^{14}\text{N}(n, p)^{14}\text{C}$  reaction, which has a much higher capture cross section). Work undertaken at Stony Brook University and Massachusetts Institute of Technology (MIT) as part of a recent United States Department of Energy (USDOE) Integrated Research Project appears to confirm that the  $^{14}\text{C}$  production from  $^{13}\text{C}$  alone will mean that future graphite waste will be classified as at least GTCC under current definitions<sup>9</sup> (Rani *et al.* 2024), but graphite manufacturers involved in the production of graphite for US SMRs continue in their efforts to reduce the initial nitrogen content of the graphite as it could ease packaging and dose concerns: the reference provides more detail on the major sources of nitrogen and how they might be reduced during the manufacturing process.

Other common isotopes found in irradiated graphite include  $^3\text{H}$  (tritium, from fuel fissions and from impurity lithium and boron),  $^{36}\text{Cl}$  (either from the purification process or absorbed from the atmosphere as HOCl), and rare earth isotopes such as europium. Residual uranium and thorium in the graphite and fuel matrix also generates fission products diffusing partially into the coolant and the primary circuit. In addition to other possible leakage of fission products from the occasional faulty fuel element, the graphite is also potentially exposed to impurities such as metal oxides transported from metallic components of the reactor circuit. Dust and volatile fractions from other parts of the reactor circuit may become trapped on the surface and in the pore structure of the graphite components and then become subject to neutron irradiation leading to further radioactive products retained in the material.

Finally, as mentioned earlier, neutron damage may affect the geometry, strength, and integrity of the graphite components, with possible consequences for the ease of dismantling, especially if intact components are to be retrieved.

Upon retrieval, most graphite waste from reactor operations will likely have  $> 4$  gigabecquerels per metric ton  $\alpha$  or  $> 12$  gigabecquerels per metric ton  $^1 \beta/\gamma$  but not heat-generating (Metcalf and Tzelepi 2019). The US classification system is essentially dependent upon the half-lives of the radionuclides that are present and their concentration, whereas graphite waste with this level of radiation would be classified as intermediate-level waste using the UK definitions. Most nations tend to plan for irradiated-waste disposal simply by category, but there is a growing realization that the

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<sup>9</sup>US waste classifications differ significantly from those applied in other countries

unique chemical, physical and mechanical properties of irradiated graphite may offer alternative strategies with both practical and financial advantages, and this has informed the international activities *via* IAEA and other bodies.

### **6.3 Dust (Contamination), Deposited Carbon, and Debris Issues**

At the time of reactor dismantling, and indeed during maintenance inspections and repair, it is probable that in addition to the blocks of the reflector/moderator, other forms of carbonaceous material may be encountered, particularly in legacy reactors. These fall into three distinct types: mobile dust which is a potential source of the spread of radioactive contamination as well as a potential interference with handling machinery, camera lenses and so forth; deposited carbonaceous material which is adherent to metal surfaces in certain zones as well as on the graphite blocks themselves (strongly dependent upon operating conditions in the reactor); and, finally, items of graphitic debris which may interact with fuel or control-rod movement and must also be accounted for when planning containment for the stack components following dismantling. These three origins are examined in turn.

#### **6.3.1 Carbonaceous Dust and Its Related Contamination**

It is important to note here that carbonaceous dust is largely confined to pebble-fueled reactor designs and is less pronounced in block-type HTGRs (Humrickhouse 2011). Carbonaceous dust producing processes do not occur in most of the legacy fission-reactor designs, and this has been confirmed during the regular maintenance inspections and decommissioning activities.

The presence of carbonaceous dust combined with metallic compound particles in the primary circuit of HTGRs may represent a significant safety issue not only during depressurization accident scenarios (Basu 2010) but also during operation, maintenance, repair, decommissioning and waste management. This is due to the high specific activity of radionuclides carried by dust particles. At the decommissioning of the Fort Saint Vrain (FSV) HTGR, the control of dust was achieved by flooding the reactor pressure vessel with water, affording the possibility of direct viewing with effective radiation shielding. At the German AVR (HTGR research reactor), the dust was fixed by filling light concrete into the complete reactor pressure vessel, which was transferred into a separate interim storage building.

Graphite dust adheres to spent fuel extracted from the core and to other graphite components like reflector blocks exchanged in periodical intervals or end of life. The dust poses challenges for the handling of these kinds of waste streams and also needs to be safely retained during transport and storage.

The amount of dust within the primary circuit significantly depends on the type of HTGR fuel elements. Block-type fuel elements generally generate less dust than pebble-bed cores with moving fuel elements within a permanent graphite reflector structure and within the pneumatically driven fuel charging and discharging systems. In the MAGNOX, UNGG and AGR reactors, with metal-clad fuel elements, dust was not observed in significant quantities.

HTGR dust safety challenges, and the needs for research and development in support of future reactors, have been intensively evaluated (Humrickhouse 2011) in the context of a US Next-Generation Nuclear Plant (NGNP).

More recent information has been obtained by investigations on the dust generated within the HTR-10 core (Peng *et al.* 2018). For example, information has been obtained on metallic and carbonaceous

dust generation by experimental simulations of abrasion of spherical fuel elements within an 8m long pebble lifting tube of the fuel supply system (Shen *et al.* 2015).

It is, however, questionable whether HTR-10 is representative of what may be observed with longer operations and other reactors, as the effective operational time has been relatively short (due to long outages) and the pebbles have not been circulated intensively. More relevant information is expected from the operations of the HTR-PM reactors.

### 6.3.2 Dust Formation and Activity

Although TRISO fuel will retain most of the fission products, certain radioisotopes will diffuse through the intact pyrolytic carbon (PyC) and silicon carbide (SiC) layers at high temperatures or will be released from defective TRISO particles. Fission reactions may occur for any free uranium and thorium in the fuel element matrix and in the graphite structures. The related fission products will diffuse within the porous matrix structure of the fuel elements. Figure 6 shows the resulting concentration profiles of the fission products Cs, Eu, Sr in the 5 mm thick fuel-free shell of a spherical fuel element (17.6% Fissions per Initial Metal Atom [FIMA], initial U-235/total heavy metal ratio = 0.17) (Moormann 2008). The  $^{131}\text{I}$  activity amounts to about 3.5 GBq/kg, which is rather high related to the total estimated  $^{131}\text{I}$  activity of  $\sim 50\text{GBq}$  outside the core region. The maximum observed specific activities in the AVR dust were  $100\text{GBq}_{\text{Cs-137}}/\text{kg}$ ,  $30\text{GBq}_{\text{Ag-100m}}/\text{kg}$  and  $400\text{GBq}_{\text{Sr-90}}/\text{kg}$ . However, such high activities cannot be expected for the actual TRISO fuel quality and due to the fact that AVR was operated with a large variety of experimental BISO (Bi-Structural Isotropic Particle Fuel) and TRISO fuel, which was stepwise improved.

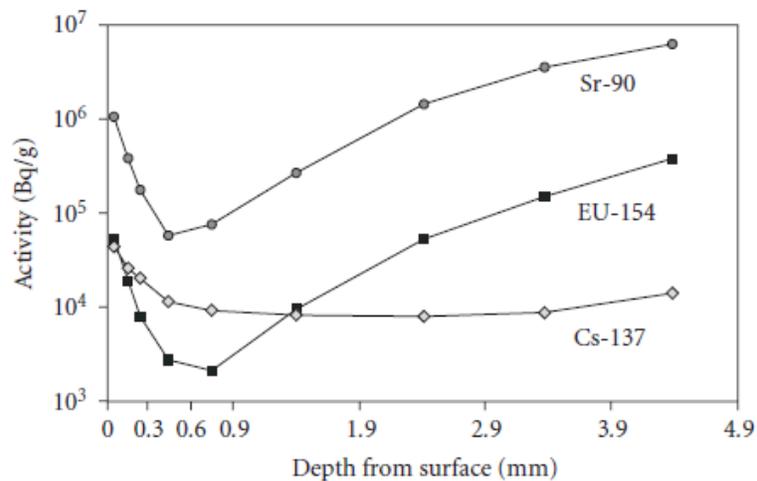


Figure 6: Concentration profiles of Cs, Eu, and Sr in the fuel-free shell of a spherical fuel element (Moormann 2008)

Dust particles strongly absorb fission and activation products, leading to enhanced specific radioactivity of the dust. The activity of  $^{14}\text{C}$  in the dust is also a significant possibility due to adsorbed nitrogen on the surface of the reloaded fresh fuel elements (see Figure 7) (Wenzel *et al.* 1979). In consequence, there is significant circulating activity borne by dust particles. A complex transport, settling, lift-off, plate-out and re-suspension phenomena dictates the radioactivity impact (Lind *et al.* 2011). Dust particles also react with the surfaces of metallic components and mobilize a considerable fraction of metallic or metal compound particles. The dust analyzed *e.g.* in the UK DRAGON (an International HTGR Prototype Project based at Winfrith, UK) reactor consisted of

only 20-25wt% of carbon material and the rest was diverse metal oxides particles (Hanson and Bolin 2007).

Additional carbonaceous material is generated via the temperature-dependent reverse Boudouard reaction, as discussed earlier. In HTRs, this is due to the presence of gaseous impurities in the helium coolant, like H<sub>2</sub>, H<sub>2</sub>O, CH<sub>4</sub>, CO, CO<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub> (Tsai *et al.* 2015) and catalytic reactions on metallic surfaces.

The influence of the presence of hydrogen in the coolant gas on the generation of dust by chemical vapor deposition has also been explained for mixtures of CO and H<sub>2</sub> (Meng *et al.* 2018). CO reacts with H<sub>2</sub> and releases H<sub>2</sub>O plus carbon particles, which are deposited and agglomerated on metallic surfaces.

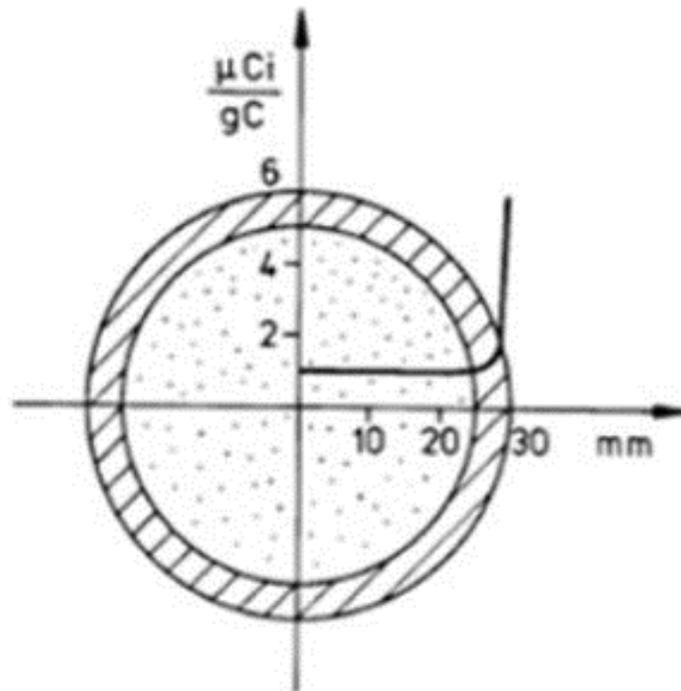


Figure 7: Radiocarbon distribution in an irradiated spherical fuel element (from Wenzel *et al.* 1979)

The reverse Boudouard reaction and the chemical vapor deposition reactions are typical for all graphite-moderated nuclear reactors but dependent on the coolant gas temperature and the kind of the coolant gas (CO<sub>2</sub> or helium). Oxidation and chemical reactions under ionizing radiation also have an impact on chemically active particles. For HTGRs, these reactions exist for both the pebble bed and for the prismatic block fueled cores.

However, most dust particles in pebble bed HTGR are generated by friction between the moving pebbles within the core and abrasion between the pebbles and the graphite reflector blocks or the metallic piping of the fuel charging/discharging system.

Figure 8 (Dietrich 2024) shows the side and bottom core reflector structures of the 300 MWe German Thorium Test Reactor (THTR-300), before operation and after shut-down and partial defueling. The scratches created by the downwards-moving fuel pebbles are clearly visible. It is evident that these

interactions create abrasion of graphite from the reflector wall and from the A3-matrix of the contacting fuel elements. Natural graphite is well known as a lubricant in air, but this is not true for nuclear graphite, which exhibits a rather high (temperature-dependent) friction coefficient under helium atmosphere. This also explains the considerable amount of carbonaceous dust observed in pebble-bed HTGR cores and in the tubing or functional elements of the fuel handling system.

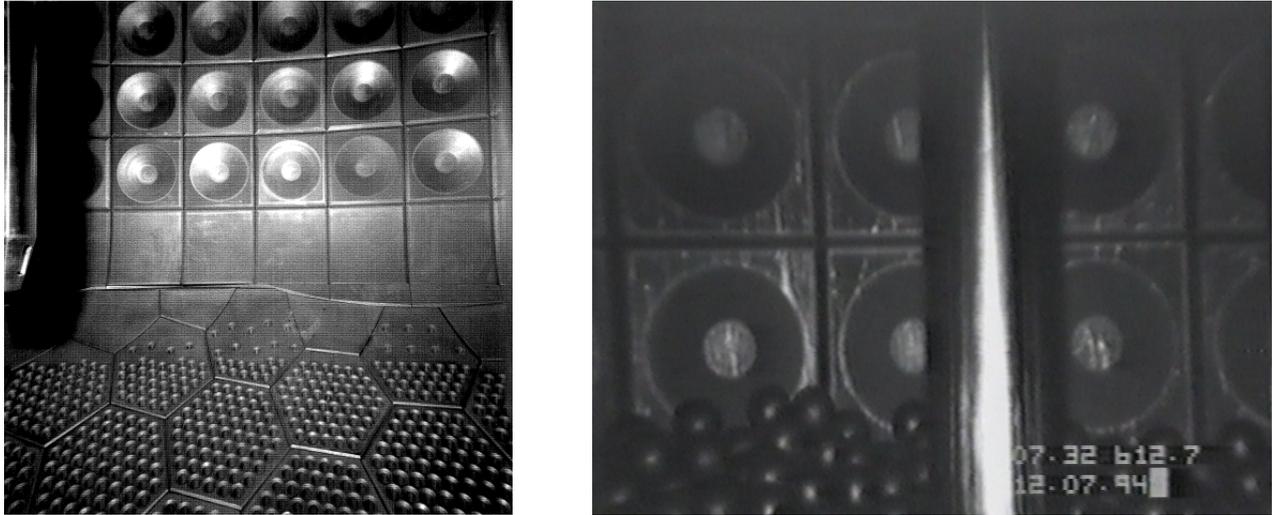


Figure 8: THTR core structures before operation (left) and after shut-down and defueling (figure courtesy of Dr. G. Dietrich, former CEO of Hochtemperatur Kernkraftwerk GmbH, (HKG))

In the German HTGR pebble-bed prototype (AVR), the accumulated amount of dust generated by abrasion is estimated to have been 50-80kg or 5kg/year (Moormann 2008). Additional carbonaceous deposition appeared due to oil ingress, as also happened at the Peach Bottom Unit 1.

The core height and the direction of the coolant flow also influence the amount of dust in pebble bed reactor systems. The AVR has a core height of 2.8m and a diameter of 3.0m, with an upward coolant flow at 1.08MPa, whereas the Chinese HTR-PM has the same diameter but a height of 11m and a downward coolant flow with a much higher pressure (7MPa) (Dong 2019). The larger core height and the downward flow with a higher-pressure drop will generate larger contact pressures on the spherical fuel elements and enhanced related abrasion rates compared to AVR and HTR-10 (1.8m in diameter and mean core height of 1.97m (Wu *et al.* 2002)).

The dust particles in the AVR had a size distribution in the micrometer range, as shown in Figure 9 (Moormann 2008) and behave like aerosols.

Dust particles measured in the HTR-10 show a similar density as measured in AVR, both about  $5\mu\text{g}/\text{m}^3$  helium coolant gas but the particle size in HTR-10 is much larger, as the dominant part is in the range of 5-10  $\mu\text{m}$ . The fission products  $^{124}\text{I}$ ,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{140}\text{La}$ ,  $^{152}\text{Eu}$  and  $^{181}\text{Hf}$  together with the solid activation products  $^{51}\text{Cr}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$ ,  $^{57}\text{Co}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$  and  $^{75}\text{Se}$ , including short-lived nuclides and long-lived nuclides, have been experimentally identified in the dust of the HTR-10.  $^{60}\text{Co}$  activity in the dust of HTR-10 is estimated in the range of  $6.5 \times 10^3 - 3.3 \times 10^4$  Bq/g. These values are much lower than the results of dust experiments  $2 \times 10^5 - 8 \times 10^6$  Bq/g in AVR (Gottaut *et al.* 1990). The same applies to  $^{137}\text{Cs}$  with  $1 - 1.5 \times 10^2$  Bq/g for HTR-10 and  $2 \times 10^6 - 9.6 \times 10^7$  for AVR. (Feng *et al.* 2017). However, this might be an effect of the short accumulated operational

period of HTR-10, the sole use of high quality TRISO fuel in the HTR-10 and the lower operational temperature compared to AVR (750°C of HTR-10 vs. 950°C of AVR).

Volatile fission products are distributed by the high-velocity coolant gas within the whole primary circuit. Visible dust layers have been observed and reported on practically all internal surfaces of the AVR (Moormann 2008), the THTR-300 (Kalinowski *et al.* 1989) and in Peach Bottom Unit 1 (Hanson *et al.* 1980). In THTR-300 the thickness of the dust layers was in the range of 5-10  $\mu\text{m}$ , despite the relative short duration of operation equivalent to only 423 full power days.

The graphite dust experiments within the AVR primary circuit revealed that the specific activity of graphite dust in the steam generator was more than 100 times higher than the dust activity on the fuel elements and more than five times that of the graphite dust in the cold helium areas (Verfondern *et al.* 2012).

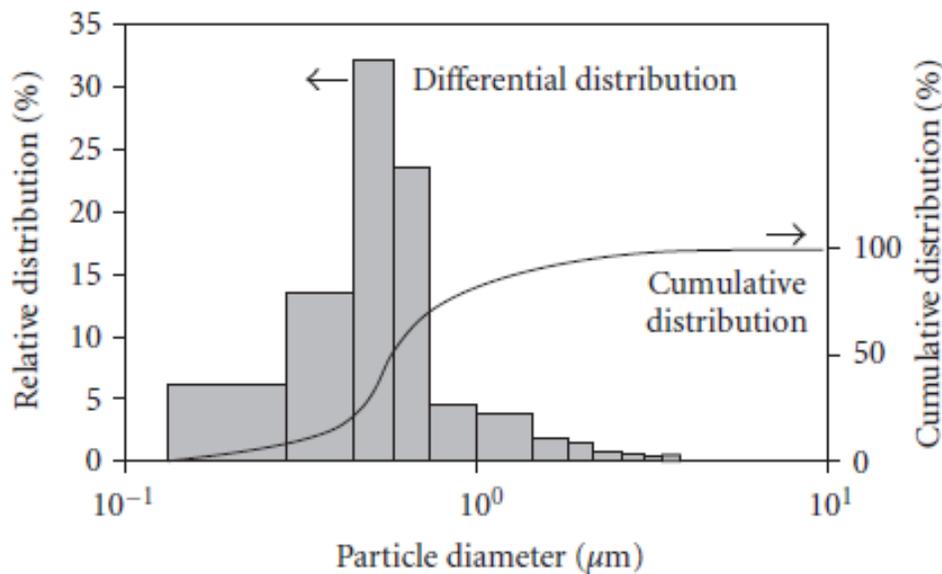


Figure 9: Weighted size distribution of AVR dust (from Moormann 2008)

Comparative sorption capabilities of nuclear graphite and of the non-graphitized binder of the A3-3 matrix have shown a much higher (1-2 orders of magnitude) adsorption of Sr and Cs on the binder material (Moormann 2008). This has a positive effect on the retention of diverse fission products in the A3-type matrix material, but it increases the activity of the dust-bound mobile activity and needs also to be considered for the treatment and reuse of separated matrix material for volume reduction purposes.

The amount of dust in block-type HTGR cores is much less due to limited abrasion effects, but it is not negligible. Non-destructive examination of 51 fuel and reflector elements from Fort St. Vrain core segment 1 (Miller and Saurwein 1980) were investigated mainly with respect to dimensional changes. These components were operated between 400-750°C and fast fluences in the range of  $0.3\text{-}1 \times 10^{25} \text{ n/m}^2$ . Visual inspection showed stains, rub marks, soot, and interface marks, due to the relative movements of the blocks against each other and the generic deposit formation processes *via* Boudouard and chemical vapor deposition reactions. The observations at FSV need to reflect the fact that FSV suffered from permanent water-ingress reducing the amount of dust. However, Peach Bottom Unit 1 experienced a steady oil ingress via the coolant blower bearings that generated carbonaceous material from the leaking oil. The Japanese HTTR is the only block-type reactor

remaining, which may – in the future – still provide some additional data on the amount of dust in the coolant gas circuit. A check of DRAGON experiences, when the reactor is dismantled, could also provide some additional evidence.

Another, but minor source, of graphite dust is relative movement of graphite components and structures against each other during heat-up due to thermal expansion and successive cool-down (Sun *et al.* 2020). Fluid-dynamically induced vibrations of fuel and reflector block columns have been observed in FSV, which also generated an intermittent clashing of the blocks.

HTR-PM will provide more relevant information after some more operational years. The HTR-PM is equipped with a separate removable sampling filter being operated in parallel to the operational dust filter in the coolant gas purification system (Xie *et al.* 2014). This will allow an easier and systematic extraction of representative dust samples from HTR-PM, being actually the only operational full-size modular HTGR with a pebble-bed core.

Overall, the considerable amount of contaminated dust circulating and partially distributed on the surfaces of discharged fuel elements or reflector blocks and on other components of the primary circuit of HTGRs represents a topic for serious safety focus (especially during depressurization accidents), and also introduces a challenge for managing, handling, storing and transporting the discharged waste. Contaminated dust will also impede the conditioning, purification and reuse of irradiated graphite or fuel element matrix material. The volatility of the contaminated dust implies a need for removal of the adherent dust directly during the discharging manipulations or for the deployment of an enclosure (contamination control), which would then prevent the release of the dust during handling, transport and storage or reactor maintenance. In the past, the operators for these operations had to wear protection suits and filtered masks to avoid any inhalation of the dust particles.

Though dust research needs were identified in Phenomena Identification and Ranking Table (PIRT) exercises, as shown in Table 2 (NRC 2008, Humrickhouse 2011), there remains little experimental and theoretical study of the topic. Recently, simulation models have been developed which may be able to contribute to study of dust circulation and deposition (Martin *et al.* 2025). It must be mentioned that the binder of the A3-matrix types behaves differently from nuclear graphite and exhibits rather different sorption behavior for fission and activation products.

There is also evidence that the dust creates deposits on diverse components within the primary circuit, which would be hard to remove and/or decontaminate (Fachinger *et al.* 2008).

### **6.3.3 Deposited Carbon on Surfaces**

Deposited carbon on surfaces has regularly been found in MAGNOX, UNGG, AGR and former HTGRs, resulting from chemical processes, and can be found on both graphite and metal surfaces. Deposits on graphite surfaces arise principally in those reactors whose coolant is carbon dioxide. In this case, the radiation-driven chemical reaction between the carbon dioxide and the graphite produces carbon monoxide, which can then polymerize as suboxide and collect on the cooler graphite and metallic surfaces, particularly in UK Magnox and French UNGG reactors. The Tokai Magnox reactor in Japan suffered severe deposition, sufficient to block coolant flow around fuel elements: this was due to a misunderstanding of the coolant chemistry and reducing the typical and already low (tens of ppm) hydrogen concentration in an effort to prevent ‘breakaway’ mild-steel oxidation, when the hydrogen was acting as a carbonaceous deposition inhibitor. In UK AGRs, where methane is deliberately added to the coolant to slow the rate of radiolytic graphite oxidation and where

temperatures are much higher, significant surface catalyzed deposition of essentially pure carbon has been found on metal-clad fuel and high-temperature metal surfaces in the steam-raising units.

The most relevant study on Magnox reactor deposits relates to a small number of samples from two reactor sites – Oldbury and Wylfa. Radiological analyses on these deposits found radioactive contamination to be ‘negligible’ (Tzelepi *et al.* 2020a), but they were found to be richer in  $^{14}\text{C}$  than the underlying graphite (Tzelepi *et al.* 2020b), which has led to the view that the probable source of deposited carbon involves nitrogen impurity which is always present at a low-level in the coolant. There are two precedents for this: formation of ‘polycyanogen’ ( $[\text{C}_2\text{N}_2]_n$ ) in the annulus-gas system of CANDU reactors when nitrogen was used as the annulus gas, and where more than 99% of the carbon present was  $^{14}\text{C}$  (Greening, 1989), and in RBMK stack graphite where the cover gas is a nitrogen/helium mixture (IAEA 2024).

Table 2: Dust Issues of the NGNP fission product PIRT (NRC, 2008)

ID	Issue (Phenomena, Process, Geometry Condition)	Importance for NGNP (High, Medium, Low)	Rationale	Level of Knowledge (High, Medium, Low)	Rationale	Status of FP Modeling (Adequate, Minor Mod, Major Need)
9	FP plate-out and dust distribution under normal operation	5–High	Starting conditions	1–Low, 4–Medium	Theory and models lack specifics	Major need
18	Sorptivity graphite	5–High	Can determine holdup and release of FP (IC and Trans.)	5–Medium	Historical data, need specific information on graphite and radiation effects	Minor mod
26	Dust generation	5–High	Vector for FP transport; possibility of high mobility	5–Medium	Limited experience; lack specific system information	Major need, import from other groups
27	(De)Absorption on dust	5–High	Provides copious surface area for FP absorption	2–Low, 3–Medium	Limited experience, lack specific details	Major need
32	Aerosol growth	1–Medium, 4–High	Low concentration growth can lead to high shape factors and unusual size distribution	5–Low	Regime has not been studied previously	Major need
33	Surface roughness	5–Medium	Affects aerosol deposition 1–5 micron particles (IC and Trans.)	5–Medium	Initial information from manufacturer; evolution of surface roughness during operation not well known	Minor mod
36	Aerosol/dust deposition	5–High	Gravitational, inertial, thermophoresis, electrostatic, diffusional, turbophoresis (Trans.)	5–Medium	Reasonably well developed theory of aerosol deposition by most mechanisms except inertial impact in complex geometries; applicability to NGNP unclear	Minor mod

Prior to the work of Tzelepi *et al.* (2020a, 2020b) mentioned above, an interesting exercise was conducted on a UK Magnox reactor, attempting a  $^{14}\text{C}$  ‘mass balance’ which included taking account of losses from coolant leakage *via* the reactor stack (Metcalf and Mills 2015). This work confirmed a suspicion that significant sources of  $^{14}\text{C}$  included the  $^{13}\text{C}$  in the circulating carbon dioxide coolant along with adventitious nitrogen in that coolant. Other work suggests that production of  $^{14}\text{C}$  from  $^{17}\text{O}$  (as  $\text{CO}_2$  and  $\text{CO}$  in Magnox systems) is also surprisingly significant in terms of production within the coolant gas and may thus explain why  $^{14}\text{C}$  appears concentrated on geometrical and pore surfaces of the graphite (Wickham and Bradbury 2010).

It is evident that such deposits may have an impact on the waste management of graphite and metallic components as well on their waste categorization in the affected systems. Enhanced  $^{14}\text{C}$  removal during oxidation processes has been experimentally confirmed by numerous research groups, suggesting the existence of  $^{14}\text{C}$  gradients within the bulk graphite which remains difficult to explain when the  $^{13}\text{C}$  source is uniformly distributed. Further, while the internal  $^{14}\text{N}$  source may not be uniformly distributed, subsequent atomic mobility through neutron collisions (damage) and recoils

might be expected to harmonize the distribution of both  $^{14}\text{N}$  and  $^{14}\text{C}$  in the bulk graphite (Vulpus *et al.* 2013, Tzelepi *et al.* 2020b).

Graphite dust also reacts with metallic components (partially by catalyzed reactions), and this can lead to fouling of heat exchanger efficiencies by deposition of dust particles and their reactions with the metallic materials (Andris *et al.* 2019). Together with particles from mechanical wear, abrasion, erosion and corrosion of metallic components, it also follows that the dust contains metallic compound particles and not only carbonaceous particles of different sizes (Lind *et al.* 2011).

The decommissioning of the German AVR prototype HTGR allowed the investigation of some piping, valves and joints, which had been removed from the primary circuit. These parts were situated between the two main blowers and the bypass for the pre-purification of the helium coolant. The helium temperature in the carbon steel pipe system was about 250 °C with 600 – 800 m<sup>3</sup>(STP)/h and helium velocities in the range of 5-13 m/sec. Loose dust was negligible within the pipes, bends and T-joint piping. The tubes were covered by a solid layer of deposits, which could only be scraped mechanically from the inner surface of pipe segments (Figure 10).



Figure 10: Removal of deposits from AVR tubing (from Fachinger *et al.* 2008)

Note that these deposits cannot have formed via the chemical processes which have already been described for Magnox and other non-HTR reactor systems. The most likely formation mechanisms are accretion of material originally present in the flowing coolant helium as particulates of aerosols, due to flow vortices, and were likely to be rich in metallic oxides arising through corrosion processes. A recent publication (Andris *et al.*, 2020) showed that *'the presence of graphite particles in the near-surface layer has a significant impact on corrosion processes due to thermally induced interactions. In this case iron and chromium are degraded in the metallic alloys, which leads to a structural change in the near-surface layer. Furthermore, the graphite particles significantly influence the formation of the oxide layers on the alloys; for example, they influence the formation speed of the layer and the layer height. The originally deposited particles thus exhibit a chemically altered composition and a different geometric shape'*.

In the AVR, activity distribution measurements indicate a dependence of the deposits thickness on the coolant flow distribution. The mechanical removal of the deposits reduced most  $^{60}\text{Co}$  but left 50% of the  $^{154}\text{Eu}$ . Figure 11 shows the distribution of radioisotopes on the sampled right-angled bend in a coolant duct (the letters E, M and A refer to sample locations, as indicated). Chemical decontamination was more effective than mechanical removal of the deposits (Fachinger *et al.* 2008).

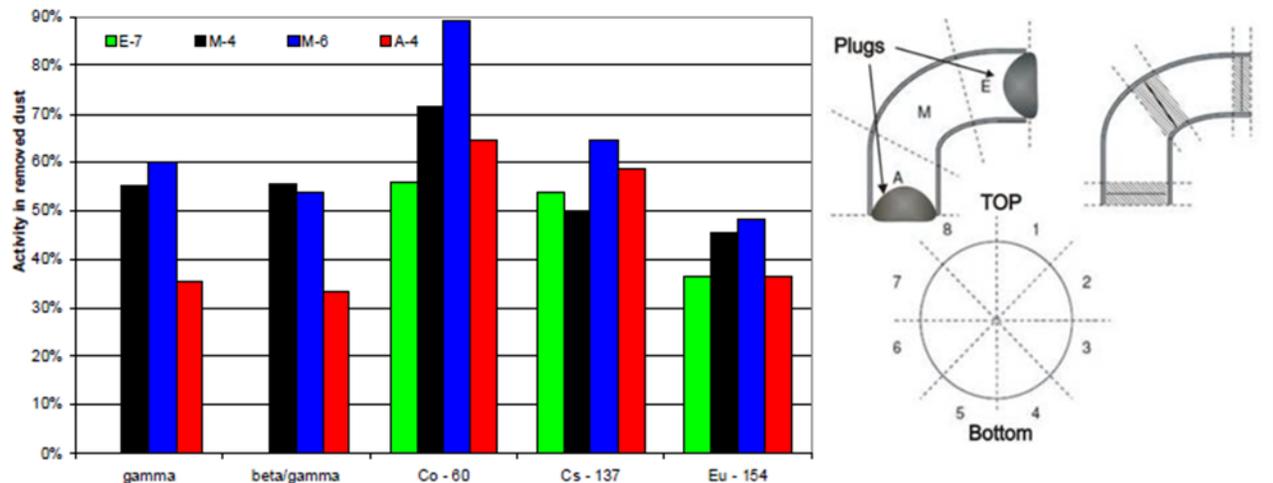


Figure 11: Fractural dose rates and activities in the scraped deposits (from Fachinger et al. 2008)

### 6.3.4 Debris

In the legacy fission reactors (Magnox and UNGG) there is very little debris present, and this has been confirmed throughout their operational lives by the regular manual and remote inspections which take place during statutory outages. UK AGRs have experienced extensive graphite-brick vertical cracking, with some evidence of fragmentation on the weaker end features of the components (Reed 2023). There is no debris information on RBMK reactors yet, except for the structural distortions already reported and where manual interventions (re-boring and so forth) may have generated some materials.

Debris from fuel elements and reflector structures have been found in HTGRs to different extents. Broken bolt heads and tie bars have, for example, been found in the DRAGON reactor, an international project located in the south of England, UK: ‘The debris on top of the reflectors was removed by introducing a ‘dustbin’, carrying sticky-headed mops, into the core cavity. The charge machine manipulated these to pick up pieces of debris and transfer them to the bin for subsequent removal from the reactor’ (DRAGON Project ‘Report 1000’, 1978).

DRAGON reactor debris appears in different forms and apparent origins. Generally, the spectrum of debris is quite large ranging from broken screws, spacers, instrumentation, insulation, graphite-keys/dowels towards damaged fuel elements and graphite components or parts of it<sup>10</sup>.

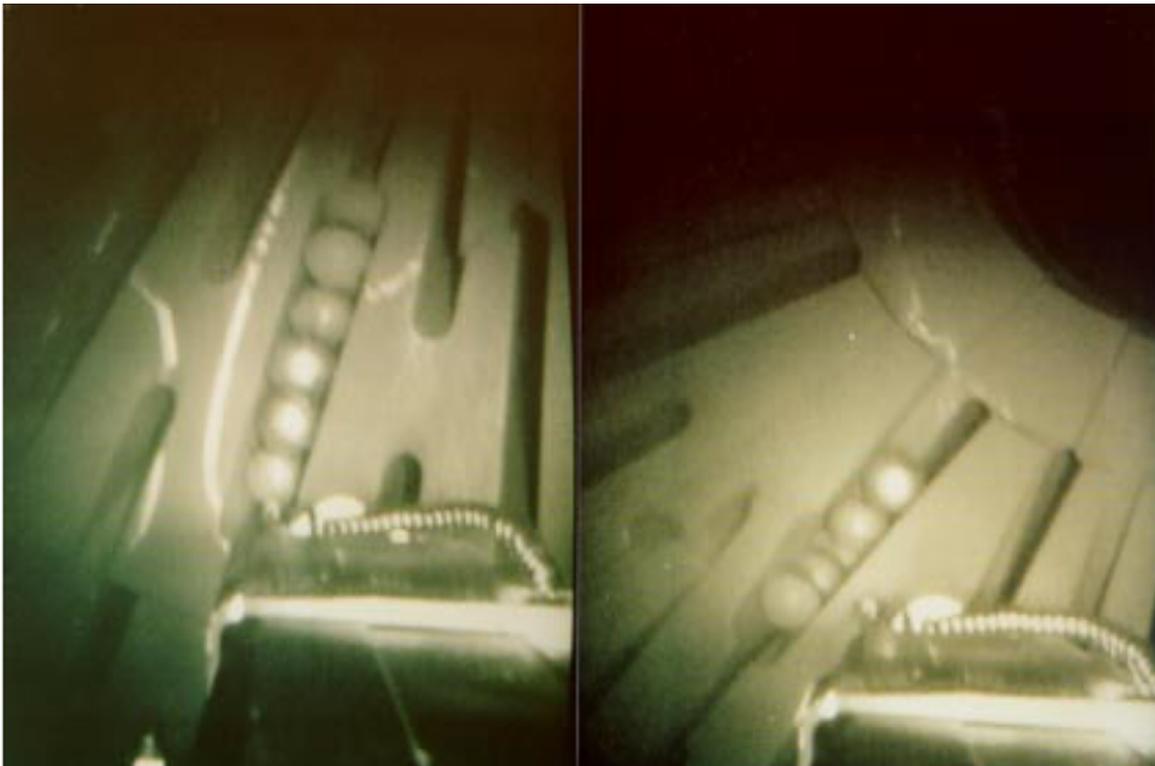
In the FSV reactor, non-destructive examination of 51 fuel and reflector elements from Segment 1 (Miller et al., 1980) has shown some small amounts of debris of the examined fuel and reflector blocks, such as damaged dowels for aligning the blocks vertically within a fuel or reflector block column. No further information was found on debris encountered during the decommissioning of FSV by the authors’ present search.

Debris from broken fuel elements is especially observed in pebble bed HTGRs. In the German AVR and THTR, broken fuel pebbles represented the dominating debris already separated and collected during operation by a separator with a rotating helix device and a collection within specific canisters

<sup>10</sup> In the context of a nuclear reactor, "foreign objects" refer to any material or debris that is not a part of the reactor's intended design or components. These objects can range from personal items like eyeglasses or tools to pieces of equipment or even debris that accidentally falls into the reactor's system.

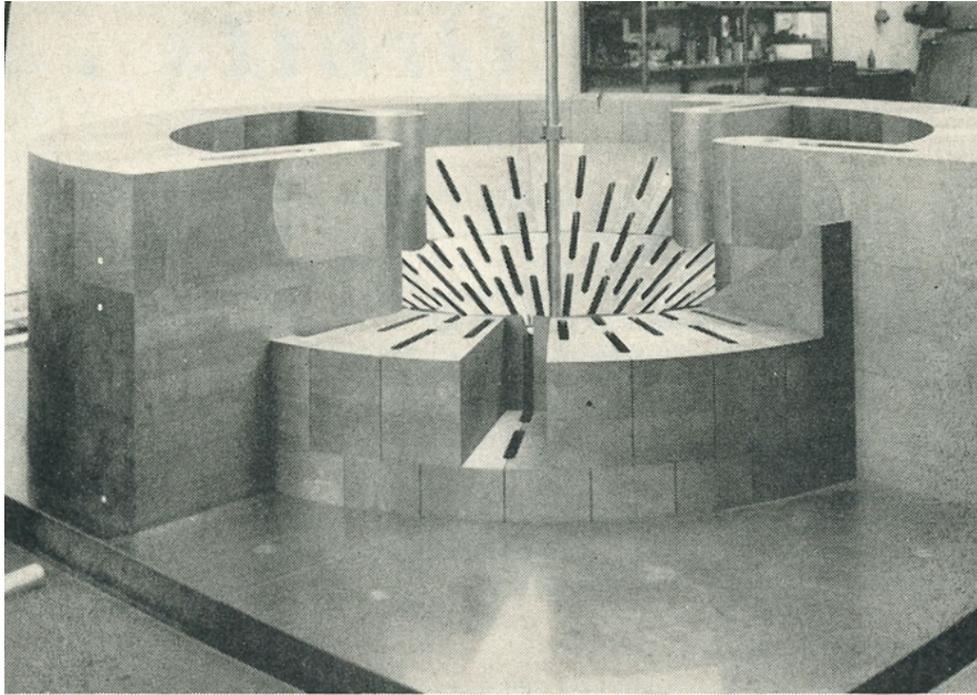
for broken fuel pebbles. Another region of broken fuel pebbles and fragmentation of parts of the graphite structure was revealed after complete defueling of both reactors.

Although a visual inspection of the upper part of the AVR core above the surface of the pebble-bed core revealed no wear or damage, the bottom reflector structure showed considerable damage of the conical graphite blocks in the area of the slit-like coolant gas exit channels. Figure 12 (Wahlen *et al.* 2000) shows that these features have become broken and widened, most likely due to fast-neutron-induced dimensional changes and differential stresses in the highly anisotropic ARS graphite, these effects being at a maximum in the cooler parts of the reactor. The widened 'slits' even captured several fuel pebbles, which should have rolled towards the fuel discharge pipe without falling into the slits if the designed width of the gas-exit channel had been maintained. The bottom reflector structure is largely broken and disarranged. A similar arrangement of coolant pathways is realized at the Chinese HTR-PM and may be chosen also for other pebble-bed reactor projects. Careful analysis of the reasons for the damage in the AVR core bottom would assist designers of future HTRs, especially as some of the currently proposed SMR designs have similar arrangements for coolant entry or exit. The captured spent fuel in the AVR bottom reflector could not be removed by manipulators and now remains in the stabilized (cemented) core, which has been transported into an interim storage building, awaiting segmentation, conditioning and packaging for final disposal. The function of the cooling channels was not significantly degraded during AVR operation because the captured pebbles were not significantly reducing the flow of the cold helium coolant gas entering the core from bottom. However, additional smaller debris items from fuel elements may have accumulated in the lower parts of the conical bottom reflector structures, which could not be inspected in detail.



*Figure 12: Cracks and residual pebbles in the AVR bottom reflector (from Wahlen et al. 2000)*

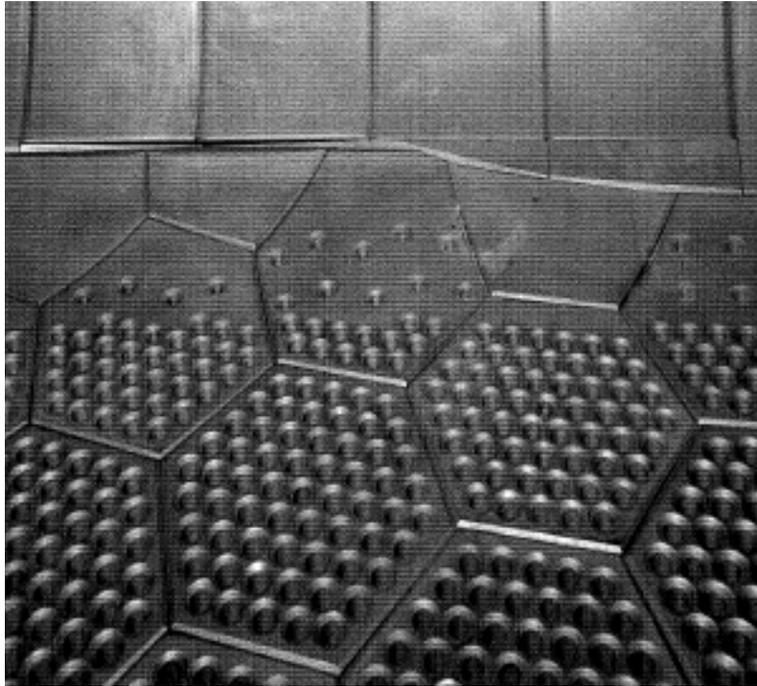
The trial assembly of the AVR core bottom is shown in Figure 13 and provides an insight into the design of the graphite structure including the protrusions (noses) for the control rods.



*Figure 13: Trial assembly of the AVR graphite core bottom structure (HTR-Archive FZJ)*

A greater quantity of fuel debris was generated in the THTR-300 reactor by the shut-down rods being directly inserted into the pebble-bed core with a pneumatic driving mechanism (in AVR they were present in four graphite protrusions as part of the side reflector and shown in the above figure). The in-core absorber rods of the THTR were intentionally inserted several times during the commissioning and generated additional fuel element debris (Baeumer 1989).

Figure 14 shows the structure of the side and bottom reflector blocks. The THTR had a different design for the coolant gas channels compared with AVR being boreholes in a hexagonal configuration with six-cm distances and a conical chamfer on the top of each borehole. This arrangement, with a distance being, by chance, identical with the diameter of the spherical fuel elements, favored a regular arrangement of the fuel pebbles on the chamfered boreholes and enhanced the forces on the movement of the pebbles.



*Figure 14: Bottom reflector of the THTR-300 (before fueling). Figure kindly provided by Dr. G. Dietrich, former CEO of HKG.*

In contrast to the AVR design, in THTR, the base of the reactor is the hottest region since the gas flow is downwards. In general, irradiation dimensional changes lessen at higher temperature.

After complete defueling, it was discovered that a large number of boreholes were plugged with small pieces of debris from broken pebbles. It can be concluded that smaller parts fell into the hot-gas chamber below the bottom reflector and are either still residing there or were transported by the helium coolant to other locations in the primary circuit.

The damaged fuel elements were sorted by two rotating helix devices and filled into specific canisters under the pre-stressed concrete reactor pressure vessel (Figure 15). However, a large proportion of the smaller fuel element debris clearly failed to be transported into the canisters for broken fuel elements during these operations and was revealed when the core was completely emptied.

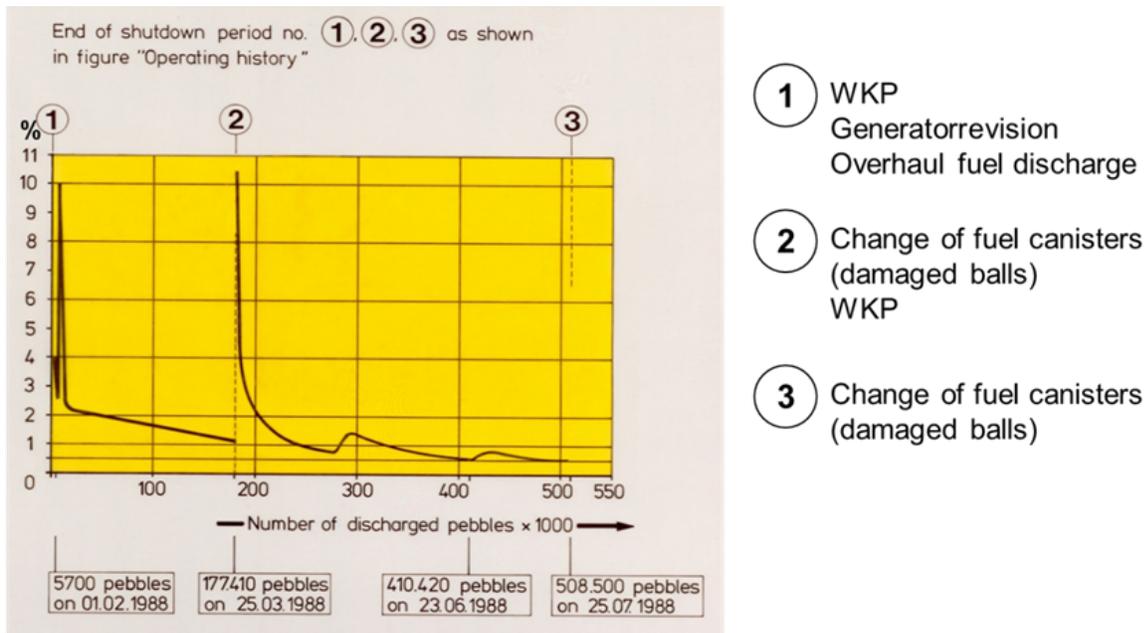


Figure 15: Discharged damaged fuel elements at the THTR-300 ('WKP' means 'Recurring Inspection'). Figure kindly provided by Dr. G. Dietrich, former CEO of HKG.

Other smaller pieces of debris were found in the gaps between adjacent blocks of the bottom reflector structure. Many gaps, e.g. around the fuel discharge pipe, were filled with debris of varying sizes, which consequently hindered the rearrangement of the ceramic core structure, e.g. after start-up and cool-down cycles, due to thermal expansion and shrinkage. This effect might have been enhanced by coolant gas bypasses through the gaps between the different graphite blocks of the bottom reflector and the fuel discharge pipe.

No significant enhancement of the coolant gas activity was observed because most of the BISO<sup>11</sup> coated particles remained intact, despite the unexpected enhanced damage rate of the fuel elements. Fuel debris was also generated within the pneumatic fuel circulation system and could have led to a jamming of intact spherical fuel elements, as seen in Figure 16:

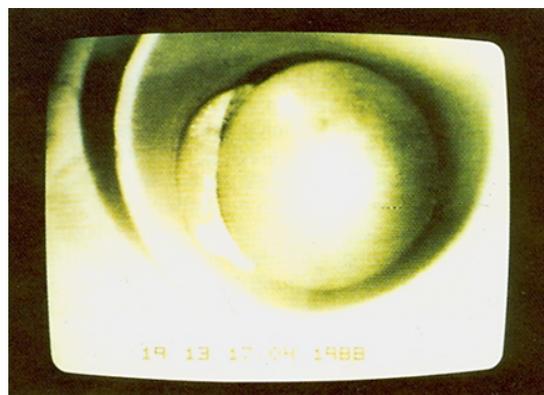
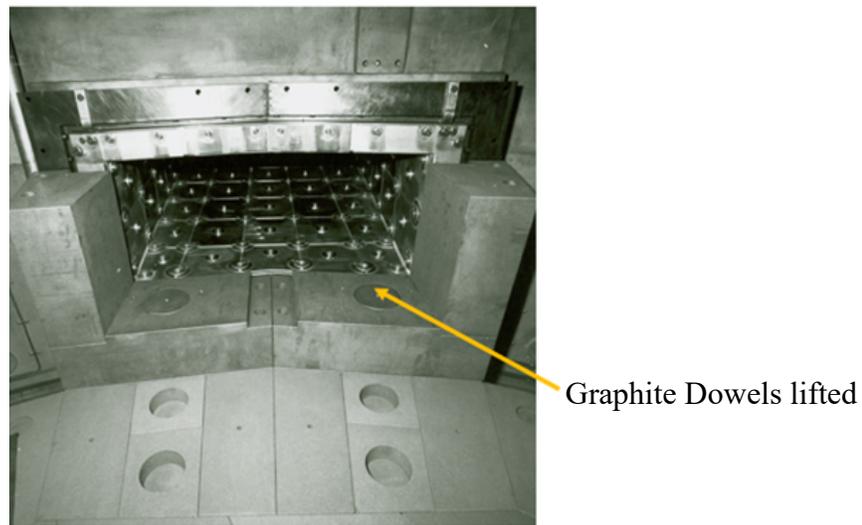


Figure 16: Jamming of fuel pebble by fuel debris (kindly provided by Dr. G. Dietrich, former CEO of HKG)

<sup>11</sup> Bistructural Isotropic fuel particles, BISO, were also employed in the Peach Bottom HTR.

It is evident that the large amount of fuel debris will affect the decommissioning of the THTR and the associated waste management including the debris and the residua of the debris distributed within the whole primary circuit. Figure 17 shows one of six hot-gas ducts and the interface with the graphite structure around the hot-gas collection chamber, during construction.



*Figure 17: Hot-gas duct and adjacent graphite structure of THTR (kindly provided by Dr. G. Dietrich, former CEO of HKG)*

During inspection at the end of the THTR operation, it was discovered that several bolts of the cover-plates in the hot-gas duct were ruptured (without impacting the function of the hot-gas duct). Some of the graphite dowels fixing the graphite structure around the entry into the hot-gas ducts were lifted by the pressure differences between the static and high-velocity coolant gas. Not all loose debris could be retrieved.

The examples show that debris should be considered for new designs with limited specific design codes and operational experience. It is plausible that future designs might include precautions for the detection and removal of debris including necessary access and repair options beginning at the design stage. The presence of debris can also affect decommissioning activities and the related waste management.

Potential debris issues are different for spherical and prismatic block fuel elements. The movement of the fuel pebbles through the core of modular HTGRs with a relatively small diameter around 3m and a large height, about 10m, result in high mechanical forces by gravity and enhanced by the pressure loss of the (down-flow) helium coolant. Due to the point contacts between the individual fuel pebbles being distributed in a stochastic arrangement between the walls of the side reflectors there is a certain probability of damage to the fuel pebbles. Thermal expansion during start-up and shrinkage of the side reflector during shut-down can induce further radial restraints within the pebble-bed core. The transport of pebbles through the fuel circulation system and the free fall on the top of the core may also create cracks, which can accumulate during the multi-refeed history of each fuel pebble. This probability is rather low but still results in few broken fuel pebbles due to the large number of circulating fuel pebbles during the operational life of the reactor. The larger parts of the broken pebbles will most probably find their way to the canisters collecting the fuel debris. The smaller parts may be captured in coolant channels or gaps between graphite components or will be carried by the coolant to dead-flow regions. This will add challenges to the repair, maintenance, decommissioning and waste management activities, especially if (damaged) TRISO fuel particles are also involved. The design of the conical bottom graphite reflector needs special attention with regards to the coolant

channels. Non-exchangeable side reflectors of pebble bed cores have to withstand accumulated high fast flux doses and might generate debris at the end of life.

For block type HTGR there is only a negligible probability that the debris contains fuel particles. This would only be the case if fuel elements are damaged during defueling and refueling activities. Small amounts of debris and wear have been observed in FSV e.g. on the dowels of the fuel elements. Other sources of debris are broken bolts, screws, spacers, metallic restraints, instrumentation, insulation material etc.

From the radiological safety perspective, the lessons to be learned from ‘prototype’ HTRs are that the behavior of the graphite at the base of the reactor, whichever direction the coolant flow is taking, needs to be carefully investigated in new designs, in order to avoid issues such as those experienced in AVR and THTR. The ability to deal with the irradiated fuel elements and, ultimately, dismantling the reactor itself, will depend upon the initial design and choice of graphite material. Candidate graphites are being investigated in materials test reactors for characterization of their irradiation behavior (the reader is referred to the numerous reports of irradiation testing undertaken principally at Idaho National Laboratory and Oak Ridge Nuclear Laboratory, as recorded in many presentations in the IAEA International Nuclear Graphite Knowledge Base). Current SMR designs with spherical fuel elements follow similar approaches for coolant pathways to those in AVR and THTR. It is plausible that future designs might consider the potential for trapping spent fuel pebbles or broken parts of fuel elements, with precluding or mitigating decisions made even at the design stage.

## **7 Planning the Decommissioning and Disposal of Graphite Waste**

There is great benefit for decommissioning and disposal planning if the entire procedure, from dismantling up to final disposal, is examined with all options considered, before embarking upon a new nuclear reactor program: this perspective has been under careful consideration for a number of years – see, for example, Wareing *et al.* (2017), Wickham *et al.* (2017). Several countries with commercial graphite-moderated reactors have already taken the decision to remove graphite as whole components with the objective of disposal in some deep underground repository in the future. This then creates a situation where, if it is subsequently decided to treat the graphite in a process which requires crumbled powdered material (such as potentially a process to reduce radioisotope content and lower the waste category, or for other reasons), then an additional complex handling and preparing operation would be needed. In this described scenario, the additional complex handling process would have been avoided if the dismantling process had involved crumbling at the time of dismantling. Another consideration is the need for ‘temporary storage’, which has different safety considerations compared to leaving the graphite in place in the original reactor vessel, and which creates additional high costs relative to direct disposal or direct processing for reuse or to simplify later stages of the disposal chain.

A holistic overview of management options for irradiated graphite, with potential future developments in the USA particularly in mind, has been published by Forsberg (2024). This paper offers a detailed review (with 64 references) of the options for management of the waste, considering the nature of the materials and their specific characteristics (e.g.  $^{14}\text{C}$  content, association with molten salt residues). The review covers the possibilities of: direct disposal to a suitable repository; recycling for reuse within the nuclear industry; and oxidation to carbon dioxide with isotopic dilution and underground sequestration. A recent publication by Wickham and Bradbury (2025) analyses the sequestration route in detail.

Figure 18 offers an example of the interconnectivity of operational stages in irradiated graphite management and their interdependence, showing the decision points and feedback loops associated with this holistic view. This diagram should be seen as complementary to Figure 2. It was created for IAEA publications to aid national authorities considering committing to procedures for individual stages, by displaying what associated procedures might follow or precede a given choice.

A number of additional points can be made which relate to the process shown in Figure 18:

- ‘Retrievable Storage’ is deliberately omitted here. If the waste can be retrieved for some future unknown ‘improved’ procedure, it is not considered as irretrievable disposal. Typically, Geological Disposal Facilities (GDFs) are designed for irretrievable disposal. Still, there can be instances of retrieval from GDFs. For example, the German facility at Asse involved corroding drums of waste in a collapsing salt mine, which was stored with other types of wastes and biological remains. A very expensive and lengthy program has been announced<sup>12</sup> to recover and move this material to another disposal environment – but no receiving disposal facility is available for these materials, yet.
- Another facility in Germany, Schacht Konrad’, (KONRAD), a former iron-ore mine intended as a repository for intermediate and low-level radioactive waste, is subject to very restrictive rules about the total isotope content which can be stored there, and these limits have not been legally amended to consider contemporary technical knowledge. Thus, the carbonaceous material from the German AVR reactor cannot be placed there because of limits on <sup>14</sup>C content, as it would require proving that the <sup>14</sup>C content was less than 1% in volatiles (Brennecke and Warnecke 1991). As already mentioned, this reactor is therefore currently laid on its side and filled with concrete in a new purpose-built enclosure pending a definite plan for its future<sup>13</sup>. There is no international consensus on acceptance criteria in disposal facilities for irradiated carbonaceous materials nor, in some instances, any opportunity to review waste-acceptance criteria based upon operational experience and improved contemporary knowledge.

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<sup>12</sup> IAEA Waste-Management Conference, November 2022

<sup>13</sup> In the long term, the carbon blocks could be disposed of in a high-level waste repository and the graphite blocks in the KONRAD facility subject to <sup>14</sup>C content.

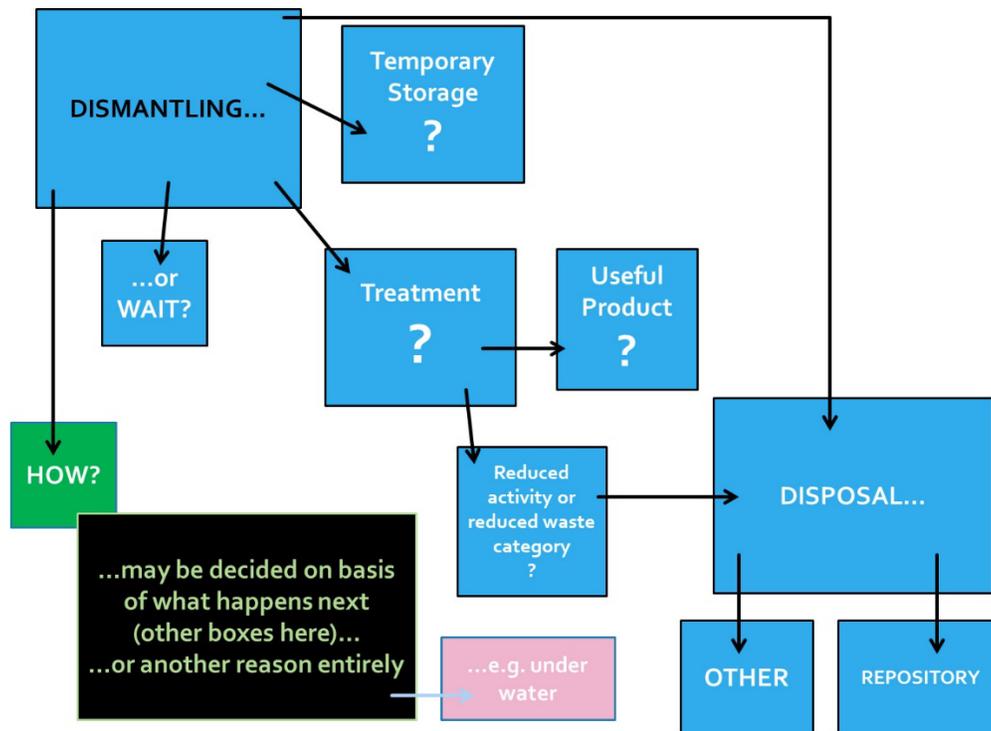


Figure 18: A holistic view of the management of irradiated graphite wastes, indicating the interactions between each potential stage of the process, created for IAEA publications to aid national authorities during decision making (from Wickham *et al.*, 2017)

A decision by French authorities to dismantle the five UNGG reactors under water was originally based on avoiding any dust explosion (which can be prevented by eliminating the requisite conditions for combustion)<sup>14</sup> (Wickham and Rahmani 2010, Minshall 2017, Antonenko *et al.* 2020)). The plan was then changed to one of using additional shielding to personnel working on the task. The decision was made without having a full analysis of the additional waste streams and costs associated with the procedure and highlights the complexity of making such decisions with incomplete information, and the potential for significant later outcomes. The plan was then abandoned when it was realized that not all of the UNGG vessels would actually be able to retain the water. A newer strategy has been adopted for dismantling in air with Chinon as the model (IAEA 2024), to gain experience for the other French reactors. If the original underwater procedure had been undertaken and subsequently followed by other countries, this would have had costly consequences (in both personnel time and monetary resources) for other nations, particularly UK, with a large

<sup>14</sup> For a dust explosion (strictly speaking, a *deflagration*) to occur, six conditions have to be simultaneously satisfied: there must be an oxidizing atmosphere; the dust material must be combustible; there must be an ignition source; the dust must be suspended (turbulent flow regime); the dust must be within a critical particle size range; and the concentration of the suspended dust must be within a critical range. In decommissioning and storage situations, it is easy to eliminate one or more of these conditions. Although the standard ISO test reveals nuclear graphite dust to be ‘weakly explosible’, it should be understood that the standard test is initiated by a very high energy firework. In reality, powdered graphite (in delaminated form) is used as a fire *extinguisher* and has been effective in remote-handling cave lines in suppressing (for example) Magnox metal fires. Excellent corroborative work has also been carried out by SoGIN in support of the decommissioning of the Latina Magnox reactor (Wickham and Bradbury 2010).

number of graphite-moderated reactors, as the reactors and infrastructure had not been built with such a plan in mind.

- It is understood that the regulators in Lithuania (VATESI) have ruled out any process other than whole-block removal for graphite from Ignalina RBMKs (the first large-scale reactors likely to be dismantled)<sup>15</sup>. No disposal facility currently exists in Lithuania, and there is therefore a plan to use a large new building for graphite storage whilst another large building for irradiated fuel storage has already been constructed (Poskas 2013). Lithuania is a country with very limited space or suitable geology for a deep repository, and a final management and disposal strategy has not yet been provided publicly.

To interpret the Figure 18 diagram for any particular situation, one first needs to fully understand the current state of the irradiated graphite in question. This ‘characterization’ covers a number of important issues, as detailed below.

### 7.1 Physical Condition of the Graphite

Before planning the dismantling of a reactor moderator or reflector, it is important to determine whether it is structurally sound or severely distorted due to fast-neutron damage (this implies in turn a knowledge of the total irradiation fluence and irradiation temperature, together with any interactions with coolants such as oxidation); also, the details of the structure should be understood and whether any other non-graphitic items are embedded within the stack.

### 7.2 Record Keeping and Loss of Knowledge

For many of the early reactors (Magnox, RBMK etc.), no quality control systems were implemented, and record keeping was haphazard. Modifications were often made as work progressed, often without any detailed recording. With the passage of time, knowledgeable people from the construction days are no longer available, and this is also true of personnel from the operational period where reactors have been shut down and left for long periods, as in the United Kingdom, where the initial plan was to leave every reactor for at least 100 years to allow significant gamma emitters to decay (such as <sup>60</sup>Co), making dismantling “easier” from the personnel dose point of view and with the possibilities of “new technology”, such as Robotics, being available when needed. Leaving the problem for future generations has short-term financial advantages but can build up longer-term problems, including potential public opposition to the policy.

A fairly recent example of complications from loss of personnel-held knowledge was the discovery that the UK reactor British Experimental Pile, Zero Energy (BEPO) had been operated as a mirror-image of the plans, after a late decision was made to exchange the ‘charge face’ and ‘discharge face’ to improve fuel temperature gradients. Only one set of plans was ever modified, and this was not the set in the formal archive. Although the reactor was shut down in 1968 and the structure sealed up, this discovery was not made until 2001 when graphite samples which had been removed in 1975 were re-visited for another purpose (IAEA 2024).

In the Magnox reactors, the realization early in the program that graphite would primarily shrink under irradiation rather than expand led to the need to insert zirconium pins between graphite blocks

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<sup>15</sup> At the time of writing, this decision is being challenged (see Section 5.2).

to stabilize columns: in the later reactors, fears that gas flows could ‘levitate’ some blocks led to the inclusion of Magnox wire. Thermocouple wiring is also threaded through the stacks. Documentation of such details could easily be overlooked when planning the dismantling.

A particular example where loss of knowledge is a major concern is the European Union funded dismantling of the Ignalina RBMKs. The second and final reactor shut down in 2009 and, since then, almost all staff who worked on the plant during operation or had detailed knowledge of the reactors are no longer available.

### 7.3 Irradiation-Induced Changes

Irradiation-induced changes can cause mechanical distortions, cracking, and fragmentation of structural materials and components. Engineers considering the dismantling of reactor stacks need to take potential distortions, cracking and fragmentation into consideration, as the forces required to lift components may be significantly greater than expected due to ‘jamming’ by debris fragments (in addition to the possibility of blocks being ‘wired together’). Differential stresses develop because the rate of shrinkage differs as one moves away from the fuel: this tends to cause ‘barreling’ of individual fuel-channel components and also, particularly found in the UK AGRs, component cracking initiated at stress-raising features such as keyways forming part of the interlinked structure. Examples of such full-length component cracking, together with the possibility of small graphite fragments breaking off and leading to difficulties with fuel movements, continue to exercise the operators and regulators of the remaining reactors: more information can be found at Zhang *et al.* (2025) and Teng *et al.* (2025). In addition, channel bores may become elliptical, and a vertical bow can develop.

In RBMK reactors, the barreling of blocks (where graphite blocks bulge in their centers after long-term radiation damage) has created a worse situation, because there were no initial gaps between graphite columns. Stresses developing in the more highly-irradiated regions have pushed out columns sideways creating a channel “bow” which became defined within Rosatom by a parameter called the “deflection arrow” – the potential sideways deflection. This becomes larger as one moves away from the center of the stack. A maximum tolerable deflection arrow of 110mm was defined, beyond which any control-rod channel would be compromised as the rod could not be fully lowered. This problem is sufficiently severe in some of the RBMKs that “re-boring” of channels has taken place alongside attempts to straighten them mechanically: It has also been reported in the international industry press (Leshchenko 2023) as recently as November 2023 that work is ongoing to ‘correct’ 137 channels in the Smolensk reactors, where the largest deflection found was 99mm. This issue is unlikely to affect the reactors at Ignalina, where operation ceased at a much lower overall irradiation.

### 7.4 The Radionuclide Content

The radioisotope content of the graphite moderator and reflector is the principal determinant of its eventual destiny and will depend strongly upon the irradiation environment and the original impurity content of the graphite. Calculations can be made which employ known parameters but rarely correspond closely to available measurements on samples taken from the stack because of the adventitious transfer of dust, debris or contamination from other parts of the reactor circuit. Such modelling was conducted for the French UNGG reactors by CEA (Commissariat à l’Énergie Atomique): there was a significant discrepancy between predicted isotope content and that found by a comprehensive sampling program from several of the reactors. The discrepancy was certainly caused, in part, by adventitious material drawn into the graphite pores by the circulating gas flow,

introducing impurity elements not originally in the graphite or adding to their initial concentrations. Whilst it could be argued that the experimental data were sufficient to inform the planning for dismantling (albeit requiring extrapolation to a large mass of material with obvious statistical uncertainties), models do exist, e.g. (Gy 1992, Heasler and Jensen 1994) which can assist in the planning of sampling programs to allow quantitative justification of the results within acceptable uncertainty criteria.

Discrepancies between calculation and measurement can result in either direction of error. For example, in a case involving the graphite reflector and thermal column of the small research reactor L-54 at Politecnico Milano, Italy, reported as part of the IAEA GRAPA Project (IAEA 2024, *Annex I*), simulations were conducted using the standard MCNP calculation methodology ('Monte Carlo N-Particle Transport') to calculate the final amount of radioactive isotopes and radiation to be expected. Where experimental data were subsequently available (for gamma emitters), they were found to be consistently higher (up to 2-3 × higher) than calculated. Measurements of beta emitters are ongoing.

Other issues which can arise include surface coverage of high concentrations of  $^{14}\text{C}$  in systems like RBMK where the graphite has been exposed to a high concentration of nitrogen. In pebble-bed systems, which generate significant levels of dust during the movement of the pebbles, characterization of the radionuclides in all carbonaceous waste forms becomes even more important.

For RBMK, the Center for Physical Science and Technology (CPST, Vilnius, Lithuania) made a significant contribution to GRAPA in regard to the Lithuanian reactors, where a major effort to identify impurities in the Russian GR-280 graphite was made, principally by neutron-activation analysis (Ancius *et al.* 2005; Bylkin *et al.* 2001, Puzas *et al.* 2010). The analysis conducted thereafter by CPST was comprehensive and thorough. Three RBMK-1500 reactor 3D models of different complexity were analyzed: a full-scale 3D model, a  $\frac{1}{4}$  3D reactor core model and a simplified 3D 4×4 fragment of the core plateau region. The possibilities of application of each model to the graphite characterization were evaluated in terms of  $k_{\text{eff}}$  convergence, computer time required for calculation of neutron activation, sensitivity analysis, and varying neutron flux (power).

The impurity data used for the RBMK-1500 reactors are shown in Table 3 for the chief radionuclides of concern (IAEA 2024). The list of radionuclides is relevant for almost all commercial graphite reactors and is shown here as a typical example. Detailed information about other reactors may be found in the IAEA references listed in this report.

The principal activation products of nuclear graphites are present (e.g. europium, iron, nickel, cobalt isotopes) along with transferred fission products (although some of those derived from uranium may well arise from impurities, since uranium, thorium and graphite are sometimes found closely allied in nature). Not shown here are considerable quantities of  $^{14}\text{C}$  which derive from the nitrogen content of the cover gas (here admixed with helium and at approximately 30% by volume in normal operation). This activated carbon will be located on geometrical surfaces of the graphite components and within transport porosity if there has been any significant gas flow through the pores. A second omission is tritium, for which there are two principal sources: fission products (about 1 in 10,000 fissions produce tritium which is highly mobile but in RBMK is likely carried away by the coolant rather than reaching the graphite as it does in a gas-cooled reactor), and lithium impurity in the graphite *via*  $^6\text{Li}(n,\alpha)^3\text{H}$  (high-energy neutrons can also produce tritium from  $^7\text{Li}$ ). Two minor production routes also exist at the high-energy end of the neutron spectrum:  $^{10}\text{B}(n,2\alpha)^3\text{H}$  and  $^{14}\text{N}(n,\gamma)[^{12}\text{C} + ^3\text{H}]$ ; the former is of little significance in graphite where boron removal has been efficient during purification, whereas the latter may have some significance with the 30% nitrogen in the RBMK cover gas.

Table 3: Experimentally Derived Impurity Data for GR-280 as used by CPST, Lithuania (IAEA 2024). Concentrations are in parts per million (ppm).

Element	Concentration, ppm	Element	Concentration, ppm
N	15±4	Zn	2.6±0.1
Cl	7±0.5	Sr	0.83±0.05
Mn	0.62±0.02	Nb	0.006±0.001
Fe	41.4±1.7	Cs	0.002±0.001
Co	0.035±0.004	Ba	1.31±0.02
Ni	0.61±0.03	Eu	0.002±0.001
Cu	0.65±0.02	U	0.0016±0.0005

Table 4 provides an example of the relative activity concentrations of uranium isotopes, fission products and activation products in irradiated graphite fuel. It is useful to cite the results of this work here since it demonstrates both what such painstaking work *can* do and what it *cannot* do. Table 4 shows the calculation for Ignalina Unit 1 in 2017 (i.e. 12 years after shutdown).

Table 4: Activity of Light Elements, Fission Products and Actinides in the Graphite Stack of the RBMK-1500 Reactor INPP Unit 1 (IAEA 2024 Annex 1). Specific activity is in becquerel (Bq) per gram.

Nuclide (decay)	2017			Nuclide (decay)	2017		
	Specific activity, Bq/g	Difference, %			Specific activity, Bq/g	Difference, %	
		Centre	Periphery			Centre	Periphery
<sup>14</sup> C (β)	6.0×10 <sup>4</sup>	14	-32	<sup>235</sup> U (α)	3.0×10 <sup>-8</sup>	3	-25
<sup>36</sup> Cl (β)	2.8×10 <sup>3</sup>	8	-23	<sup>236</sup> U (α)	2.1×10 <sup>-6</sup>	2	-4
<sup>54</sup> Mn (γ)	4.8×10 <sup>-1</sup>	11	-43	<sup>238</sup> U (α)	6.9×10 <sup>-6</sup>	-6	21
<sup>55</sup> Fe (γ)	6.1×10 <sup>4</sup>	12	-29	<sup>237</sup> Np (α)	3.7×10 <sup>-6</sup>	0	-12
<sup>60</sup> Co (γ)	4.4×10 <sup>4</sup>	8	-23	<sup>238</sup> Pu (α)	3.0×10 <sup>-1</sup>	-14	26
<sup>59</sup> Ni (γ)	37	4	-14	<sup>239</sup> Pu (α)	1.7×10 <sup>-2</sup>	-7	6
<sup>63</sup> Ni (γ)	6.0×10 <sup>3</sup>	9	-25	<sup>240</sup> Pu (α)	6.3×10 <sup>-2</sup>	2	-11
<sup>65</sup> Zn (γ)	9.8×10 <sup>-3</sup>	27	-52	<sup>241</sup> Pu (β)	6.7	-6	2
<sup>90</sup> Sr (β)	5.4	11	-36	<sup>242</sup> Pu (α)	1.6×10 <sup>-3</sup>		
<sup>106</sup> Ru (γ)	4.0	7	-30	<sup>241</sup> Am (α)	1.9×10 <sup>-1</sup>	-7	7
<sup>109</sup> Cd (γ)	1.6×10 <sup>-2</sup>	16	-45	<sup>242</sup> Am (α)	5.2×10 <sup>-4</sup>	-19	59
<sup>125</sup> Sb (γ)	38	12	-38	<sup>242m</sup> Am (α)	5.2×10 <sup>-4</sup>	-19	59
<sup>133</sup> Ba (γ)	372	10	-32	<sup>243</sup> Am (α)	4.1×10 <sup>-2</sup>	4	-35
<sup>134</sup> Cs (γ)	99	2	-15	<sup>242</sup> Cm (α)	4.3×10 <sup>-4</sup>	-19	59
<sup>137</sup> Cs (γ)	19	11	-36	<sup>243</sup> Cm (α)	3.6×10 <sup>-3</sup>	-7	-4
<sup>152</sup> Eu (γ)	5.3×10 <sup>-5</sup>	-10	12	<sup>244</sup> Cm (α)	12	19	-63
<sup>154</sup> Eu (γ)	9.3	-4	61	<sup>245</sup> Cm (α)	1.6×10 <sup>-3</sup>	20	-69
<sup>155</sup> Eu (γ)	1.4	-4	65	<sup>246</sup> Cm (α)	3.3×10 <sup>-3</sup>	45	-83

Tritium content and behavior during dismantling and handling of the waste is of great importance from the personnel radiation dose and contamination points of view, and it is normally of concern during the operational phase of a repository (half-life 12.3y). A very detailed survey of the issues around tritium is given by Wickham and Bradbury (2012).

Generally, tritium is also released by absorber systems if not retained in specific TRISO coatings surrounding a neutron absorbing kernel (e.g. boron carbide). This approach has been applied at the German THTR by absorber pebbles containing boron carbide BISO particles. The two weak beta emitters which are perceived to be of the greatest concern are <sup>36</sup>Cl and <sup>14</sup>C, primarily because of their long half-lives (301,000 and 5,760 years respectively). Long half-lives equate with very low disintegration rates (Curies or Becquerels) and therefore a hazard can only be created if these isotopes exist in a chemical form and under specific conditions where they can be concentrated sufficiently to give a meaningful exposure. Nonetheless, there may be a public perception that long half-life is, in itself, a consideration for safety. In various planned or existing waste facilities internationally, limits on the total content of weak beta emitters have sometimes been prescribed by

the relevant authorities. The limits chosen by other countries are sometimes lower relative to those prescribed by U.S. policies, which involve risk-based technical assessments.

The chemical form in which these isotopes will exist in graphite waste impacts directly upon potential volatility, solubility during any leaching event, and are important for determining the overall quantities of a particular isotope which are acceptable to a repository facility under its radioisotope acceptance criteria. A valuable example of the significance of chemical form is  $^{36}\text{Cl}$  in graphite. Concerns about this isotope have varied widely between different organizations internationally. For a long time, France's regulators put more emphasis on  $^{36}\text{Cl}$  than  $^{14}\text{C}$ , until it was found that assays of this isotope in irradiated graphite had been seriously overestimated. Additionally, and perhaps more importantly, research at The Institute of Physics of Lyon, (Université Claude Bernard Lyon 1 (INPL)) using implanted  $^{37}\text{Cl}$  in a study employing XANES (X-Ray Absorption Near-Edge Structure analysis) showed that the chlorine remaining in irradiated graphite was largely of organic form, whilst inorganic chloride was largely lost during reactor operation (Vaudey *et al.* 2010). However, there is some ambiguity when other results are taken into consideration, and it remains possible that some proportion of chloride may result especially in graphite irradiated at lower temperatures: the situation is explained in detail in Wickham and Bradbury (2012) which includes consideration of the recoil effects when the  $^{36}\text{Cl}$  isotope is initially formed and its potential movement around the graphite structure. There have been some reports of 'clustering' of the isotope in localized regions of the irradiated graphite, most recently in the context of RBMK graphite (Pavliuk 2024).

The Wickham and Bradbury (2012) document currently remains the most comprehensive review on all aspects of  $^{36}\text{Cl}$  formation and behavior in irradiated graphite, including such leaching tests which have been reported and a discussion about its behavior should it reach the geosphere of biosphere. In this latter respect, it should be noted that all commentary on this latter issue appears to rely on a key reference by Sheppard *et al.* (1996), in which all predicted leaching behavior is predicated on the assumption that the  $^{36}\text{Cl}$  is in the form of inorganic chloride. Given that the majority of evidence reports that residual  $^{36}\text{Cl}$  in irradiated graphite will be in the form of covalent C-Cl bonded entities ('organic form'), care must be taken in applying any of the older studies to new situations under investigation.

Turning to  $^{14}\text{C}$ , we again encounter the importance of chemical form – carbonate (carbon monoxide and carbon dioxide in gaseous form) being the most likely inorganic compounds, while the organic form can potentially manifest as hydrocarbons (methane etc.) or more complex molecules (the pitch used in graphite manufacture is a rich source of organics and contains quinolines, which can contribute residual chlorine atoms as well). Again, the concerns rest upon leakage from repositories and storage facilities, with gaseous  $^{14}\text{CO}_2$  and any  $^{14}\text{CO}$  which might be present potentially accessing spaces to which humans or other biota have access and might inhale. Escape into the geosphere potentially stabilizes this as solid carbonate species in the rocks (as in current deliberate carbon capture and storage activities), but organic forms and  $^{14}\text{CO}$  can potentially persist and get into the biosphere – e.g. plant life etc.

Wickham and Bradbury (2010) review evidence on  $^{14}\text{C}$  in graphite waste sources, its formation, and available data from numerous reactor investigations. As previously explained, formation from  $^{13}\text{C}$  is unavoidable as the stable isotope constitutes 1.1% of natural carbon; efforts are currently being made

to limit the amount of nitrogen in the materials of graphite manufacture<sup>16</sup> to mitigate the formation from <sup>14</sup>N, although exposure to nitrogen during production, machining and during reactor construction and operation (e.g. impurity in helium coolant of HTRs or introduced with the fuel pebbles) will produce <sup>14</sup>C during irradiation which may be distributed on geometrical or pore surfaces rather than randomly – a prime example of the latter is the RBMK design which is irradiated under a cover gas containing between 30%-70% nitrogen.

There is an additional and important issue applying to <sup>14</sup>C in graphite. Work initially introduced at the Jülich research center in Germany and then taken up by other research groups, notably Boise State University in the USA, CEA in France and The University of Manchester in the UK, has shown that by heating the graphite in a mildly oxidizing atmosphere, the initially formed CO/CO<sub>2</sub> is enriched in <sup>14</sup>C compared with the average specific activity of the graphite sample. This has a twofold advantage – the potential to reduce the waste category, and also the potential to recover <sup>14</sup>C which is a valuable medical resource world-wide. Worth *et al.* (2017) detail the most recent research and offers extensive references to the earlier work. Earlier work has shown that some <sup>14</sup>C can be released in an advantageous ratio with <sup>12</sup>C when heated in an inert atmosphere, suggesting that the <sup>14</sup>C in this case resides in a labile form on the graphite pore surfaces (Podruzhina 2005).

A useful summary of decontamination options for irradiated graphite, prepared in support of HTR-PM, is given by Fu *et al.* (2022).

For future HTR and MSR, we must also consider the potential for fission products and heavy isotopes to enter the graphite structures from occasional fuel failures.

## 7.5 Wigner Energy

A clear understanding of Wigner energy has been derived over the last 67 years or so (post-Windscale accident). Wigner energy content, which will be dependent upon the irradiation environment, is unlikely to be a significant issue in the new reactor designs being proposed, subject to verification of temperatures at the extremities of the reflector stack where some small accumulation may be encountered.

When considering the implications of Wigner energy accumulation, it is important to understand that it is not the *total* stored energy which is necessarily important, but the rate at which it can be released (as a function of temperature rise, which has the same units as specific heat capacity and therefore determines how much external heating the graphite can tolerate without the risk of an uncontrolled further release of energy). In exceptional cases, graphite with a potentially high rate of release of Wigner energy may require specific handling and management procedures: an example of this is control-rod displacers from RBMK which experience very high fluence but at relatively low temperature (~70°C) (Pavliuk *et al.* 2019). Thermal transport through bulk graphite components has been studied in detail in the specific context of dealing with the RBMK control-rod displacer graphite (Pavliuk *et al.* 2022). In many cases, such as the lower graphite layers in UK Magnox reactors, small

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<sup>16</sup> Manufacturers in USA and Japan are undertaking these studies but currently the information is proprietary and there is no published reference

quantities of Wigner energy can exist which have saturated<sup>17</sup> and do not represent a hazard in management.

A concern was raised by potential operators of the geological disposal facility planned for the UK about releases from UK Magnox reactor graphite at miniscule rates, lying well below the limits of detection of modern laboratory instrumentation. The concern was about two differing scenarios: firstly that if the graphite is stored in what amounts to an adiabatic environment (closely packed, other minor heat-generating waste adjacent), could temperatures eventual rise to a point at which an uncontrolled release could occur?; secondly, what if some kind of fire started in the repository and the graphite was heated up for the first time since disposal? It is known that measurements on Windscale Pile 1 graphite in 1958 and then again in 1998 showed no change in release rate behavior for comparable samples, which is extremely encouraging and entirely in line with expectation. It is also interesting that graphite samples recovered from Windscale Pile quite close to the fire-affected zone have retained their Wigner energy despite a quasi-adiabatic environment. The Wigner energy data for the Brookhaven Graphite Research Reactor material, verified by one of the present authors and comparable with that from Windscale, was used to justify ‘direct demolition’ of the reactor-graphite stack by (literally) smashing it into smaller pieces and heaving those pieces into modest waste containers for transport on public highways to suitable holding sites for temporary storage.

One of the best and most comprehensive recent studies of Wigner energy to support reactor dismantling has come from the Indian reactor CIRUS as part of the IAEA GRAPA program (IAEA 2024, *Annex 1*), where the reflector graphite and thermal column have been extensively sampled and subjected to differential thermal analysis. One sample did exhibit a release rate exceeding the specific heat capacity of the graphite over a small temperature range, but no special precautions are needed to handle the material under normal working temperatures.

In the specific context of HTGR, it is potentially beneficial not to irradiate graphite at temperatures below 250°C as to minimize accumulation of Wigner energy. A US review of stored energy release of irradiated graphite (Gallego and Burchell 2011) came to the following conclusions:

- The majority of the experimental data available on studies of stored energy and release of stored energy date back to the 1950s and 1960s.
- The total stored energy in irradiated graphite, for a given irradiation temperature, increases with irradiation dose and appears to reach a saturation level with increasing irradiation dose.
- The rate accumulation of stored energy, as well as the saturation level, decreases with increasing irradiation temperature. Actual experimental data to support the trend is available for temperatures up to 450 °C and irradiation doses <2 dpa.
- The release of stored energy exhibits a characteristic peak at around 200°C for samples irradiated at low temperature; the peak will shift to higher temperatures, and the magnitude of the release will diminish significantly as the irradiation temperature increases.

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<sup>17</sup> ‘Saturation’ means that the rate of formation of atomic displacements has been matched by the combined effects of thermal and irradiation annealing.

- There is some evidence of residual stored energy on samples annealed at temperatures up to  $\sim 1000$  °C, indicating that higher temperatures are required to release the stored energy.
- Additional evidence was found, for graphite irradiated at a low temperature, that a second release peak is possible at temperatures between 1200 and 1500 °C (see Figure 19).

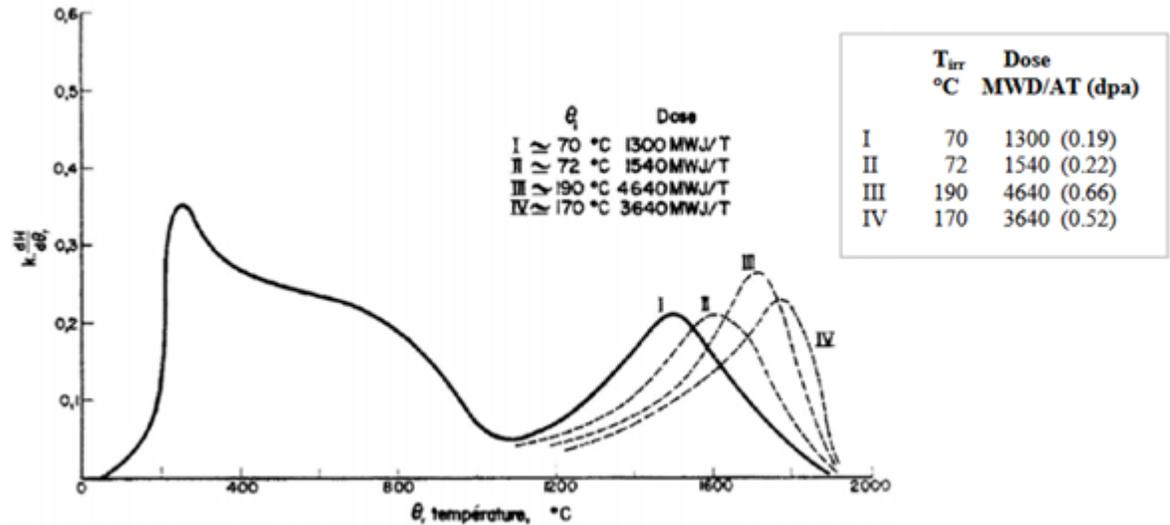


Fig.19: Nature of Wigner energy release-rate data from the BR-2 reactor graphite (from Gallego and Burchell 2011)

- The Wigner energy releases at high temperatures ( $> 1200$ °C) can be explained by the annealing of more complex defect structures and will be more relevant for core heat-up accident scenarios than for waste management.

Note that any form of thermal treatment ahead of eventual disposal will remove the perceived Wigner energy threat entirely.

## 7.6 Dismantling Approaches

It is often assumed that graphite components will be removed from existing reactors in a reversal of the process used to build the stack (e.g. removal as intact blocks). Removal of graphite components involves considerable resources in engineering equipment and shielding, and may be compromised by distortions, cracking and (in oxidizing coolants) weakening of the structure through irradiation damage by neutrons.

Considerable work has been undertaken in pursuit of this goal, most significantly by the French in support of Chinon where the strategy for dismantling the UNGG reactors is presented comprehensively in the ANDRA (Agence Nationale pour la Gestion des Déchets Radioactifs, France) ‘Graphite Reference Guide State of Knowledge’ (ANDRA 2015) which also contains a comprehensive account of all types of characterization data from the French reactors. Appropriate pilot-scale mechanical grabbing equipment has been developed with further guidance coming from EdF (Electricité de France) and CEA. Considerable design work was also undertaken by UKAEA on behalf of Società Gestione Impianti Nucleari, Italy (SoGIN) for the Latina reactor. SoGIN later embarked on an independent and dedicated design program which is comprehensively described in IAEA (2024) (Annex 2). A further Italian development has come from an engineering group at

Politecnico Milano where improved suction-cup lifting equipment has been demonstrated (Chebac *et al.* 2025).

Two very significant examples of successful whole-block removals relate to prototype reactors with relatively low cumulative irradiation. The first, at FSV, saw intact hexagonal graphite components (fueled and unfueled) lifted into containers beneath shielding water, but one should note that some irradiation damage was evident because the pre-formed screw threads associated with the blocks to enable this process had become deformed and some difficulties were encountered (Fisher 1998). The flasks holding this material remain in an ‘indefinite’ holding facility elsewhere in the U.S. (USA has a number of such facilities such as the Hanford Site, remote regions of Nevada, *etc.*).

The second example is the Windscale prototype AGR in the UK. This was the ‘demonstrator reactor’ for the 14 full-sized commercial AGRs which followed, eight of which currently remain in operation. The blocks of graphite (in this prototype, PGA rather than Gilsonite) were retrieved intact (Figure 20), placed in a suitable container for extraction through the roof of the building, placed in concrete storage containers and are now held in a temporary storage facility on the same site. This was an important demonstration that complex graphite structures could be dismantled, although the total irradiation was very much lower than in a commercial electricity-generating plant (Halliwell 2012). As already mentioned, later experience from more highly irradiated stacks shows the value of developing a simpler methodology where there is potential for component distortion and jamming by dislodged graphite fragments.

Figures 21a and 21b shows equipment designed and tested in the UK for ‘crumbling’ graphite along with a vacuum system for its removal to suitable containers - a process colloquially known as ‘Nibble and Vacuum’. This is an especially useful tool in situations where the exact content of the graphite stack is unknown and may contain items such as zirconium pinning of the blocks, embedded thermocouple wires, and so forth, and obviously valuable if a subsequent treatment requiring crumbled material is envisaged. It also offers the potential for greatly simplified engineering using remote rovers and also offers an option for greatly reducing personnel dose exposure.



Figure 20. Removal of entire graphite blocks from the Windscale Advanced Gas-Cooled Reactor prototype. (Courtesy BNFL (British Nuclear Fuels Ltd.) UK)



Figure 21: Nibbling tool (left panel) and 'vacuuming' equipment (right panel) developed as an alternative procedure for the removal of graphite from reactor cores. (Courtesy of GNAT, UK)

It is important to record some additional dismantling successes, albeit covering only the first stage of the disposal process and, in one case, *not* involving whole-block disposal. Each gives useful insights to guide future activities, elsewhere:

- GLEPP (Graphite Low-Energy Experimental Pile). This was a small graphite stack on the Harwell site in the UK used primarily for reactor-physics experiments. It had such a low overall irradiation that it could be dismantled essentially by hand. Because the  $^{14}\text{C}$  and  $^3\text{H}$  content in the graphite was so low, it was crumbled (subsequent to dismantling) and then

oxidized in an industrial incinerator alongside other materials, with no radioactivity controls needed<sup>18</sup>.

- BGRR. The Brookhaven Graphite Research Reactor on Long Island, USA was operated at low temperatures and commenced operation in the early 1950s. Following the infamous fire in a similar facility at Windscale UK in 1957 (Arnold 1992), great concern was expressed that a similar build-up of Wigner energy was occurring in the BGRR and that a similar event might occur. There followed an anxious period of research during which the fundamentals of the oxidation in air of nuclear graphite were fully investigated and this legacy led to continued safe operation for many years. As noted earlier, one of the present authors (AJW) was responsible for interpretation of Wigner energy data for BGRR ahead of the final dismantling, which adopted a very simplistic approach – a large ‘excavator bucket’ inside a suitable temporary contamination containment, loading graphite debris into skips which left the site by road transport, again for a holding area. All traces of the reactor have been removed from its original building (Kirby 2011). In contrast, the UK Windscale piles remain largely untouched, though thorough investigations of these regions were undertaken in the late 1990s, including photography and the recovery of graphite samples by UKAEA which demonstrated that dismantling could safely proceed: however, no further action has yet been taken.
- ADE-5. Removal of graphite blocks from this Russian production reactor began recently and was reported as part of the ‘GRAPA’ project (IAEA 2024 *Annex 2*). A recent paper (Pavliuk *et al.* 2023) gives a comprehensive and useful account of dismantling and pre-dismantling issues encountered in the graphite stacks of a number of Russian reactors.
- The Danish materials testing reactor DR-3 (Danish ‘DIDO’ pool-type research reactor), with a graphite block reflector and similar to ‘DIDO-type’ heavy-water moderated graphite reflector research reactors in the UK, Germany and elsewhere, has been completely dismantled and the irradiated graphite taken to storage alongside the thermal column material from the pool-type reactor DR-2 (Dansk Decommissioning 2018).
- A final example, albeit not conventional dismantling, is seen as a permanent solution for another Russian production reactor, in the ‘GRAPA’ project. This reactor, constructed below ground level, has been ‘entombed’ after stabilization with an epoxy-based filler and sealed off, with the removal of all buildings, effectively returning the site to brownfield status. Such ‘*in-situ* disposal’ would not likely be approved by most national regulatory authorities and also could only be achieved in this very special situation.

## 8 Potential Treatments for Graphite Waste

### 8.1 Why Treatments May be Considered

Even if there were an immediate option to deploy the graphite waste, suitably stabilized and packaged, to a final repository, licensees may choose treatment of the waste for economic and safety

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<sup>18</sup> The local authorities became concerned about graphite fire in the stored, crumbled material: graphite does not burn as such below ~3700K, although the chemical oxidation process in air achieves a measurable rate at around 670K. See Paul *et al.* (2023)

reasons. For cases where irradiated graphite is in temporary storage or remaining in reactor with final destiny remaining unclear (the majority of cases), some licensees may consider potential treatments of irradiated graphite waste to the following reasons:

- Reducing the volume of material to be managed;
- Reducing the waste category, with consequent financial savings and simplification of handling, including potential waste management at alternate facility choices;
- Obtaining one or more useful isotopes for medical use or other activities;
- Improving retention of radionuclides during storage and final disposal;
- Recycling into future nuclear activities (*via* reprocessing).

A modest delay in removal and handling, to allow short-lived (mainly gamma-emitting) radionuclides to decay, could also be utilized to reduce radioactivity. For example,  $^{60}\text{Co}$  will have decayed in just under 55 years (ten half-lives), tritium (which also presents significant risks to personnel) will remain an issue for 150 years. Long-lived beta emitters,  $^{36}\text{Cl}$  and particularly  $^{14}\text{C}$ , will not decay significantly. These radionuclides are often the subject of comments by public stakeholders, even for cases where contemporary calculations show no subjective risk.<sup>19</sup> In the already operable LILW repository at the Loviisa site in Finland,  $^{14}\text{C}$  and  $^{59}\text{Ni}$  isotopes dominate the long-term safety analyses (see Nummi 2019, fig. 2-1, page 21).

Delaying dismantling can reduce personnel doses or risks of personnel doses. However, a serious downside to delaying dismantling in order to wait for accessibility to become easier, is that there can be great loss of knowledge about the reactor construction, especially for older plants where quality assurance and record keeping were minimal. An example of this has already been given, in the context of the UK experimental reactor BEPO.

## 8.2 Treatment Options

Some examples of various treatment initiatives are now given, for which further information will be found in the principal IAEA references:

Reducing overall activity by removing isotopes. This may be both to attempt to lower the waste category, and because certain isotopes needed for medical and other purposes are in short supply. Where the isotope is not collected for such future usage, processes have been developed to concentrate radioisotopes into a small amount of residual waste, to be disposed separately from the main bulk of the graphite. A prime example of this is thermal treatment under mildly oxidizing conditions to yield CO/CO<sub>2</sub> off-gas which is enriched in  $^{14}\text{C}$  compared with the specific activity of the residual graphite. This has been investigated in a number of laboratories as elaborated earlier (see Worth *et al.* 2021 and references therein) and is particularly successful where there is an enhanced surface covering of deposits and/or where adventitious nitrogen, subsequently converted to  $^{14}\text{C}$ , has been collected. RBMK core graphite is an ideal example of this, but most studies so far have been conducted using graphite from UK Magnox reactors and the BEPO reactor. There remain aspects of the production and subsequent mobility of  $^{14}\text{C}$  from  $^{13}\text{C}$  which remain to be fully explained and an analysis of recoil behavior of the  $^{14}\text{C}$  atoms following formation, alongside atomic displacements of all carbon atoms driven by fast-neutron collisions, is under consideration. Tritium can also be removed by thermal treatment in an inert atmosphere at around 450°C, as recently demonstrated on

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<sup>19</sup> ‘Subjective risk’ is a perceived risk. Technical recommendations should be based upon ‘objective risk’ – in other words, analyzed on the basis of data and technical knowledge.

graphite and carbon-fiber reinforced first-wall tiles recovered from the JET torus (Wickham *et al.* 2022): this is of importance since continuous production of tritium is necessary (to counter its decay) to support future fusion programs such as ITER, and for military use, and the UKAEA is currently considering the economics of recovery from fission-graphite sources (Reynolds 2025). Nearly complete removal of tritium in irradiated graphite has been observed at 1300°C under inert atmosphere. Repeated exposure and subsequent heat treatment at 900°C under nitrogen atmosphere also showed an efficient reduction of  $^{14}\text{C}$  (Vulpus *et al.* 2013). This process generates only a small amount of secondary waste (few percent of carbon loss).

Heating (in its own right) may be undertaken under controlled conditions to anneal Wigner energy to an appropriate degree. Modest heating to 450-600°C will remove a high proportion of such energy in graphite sources which have been irradiated at relatively low temperatures, greatly reducing the risk of spontaneous releases in the later stages of waste management in the rare cases where this is possible.

Incineration in a fluidized bed has been demonstrated by Framatome (Guiroy, 1996) and more recently by Russia (Girke *et al.* 2012), but obviously leads to the release of  $^{14}\text{C}$  to the atmosphere. Against the background of cosmic-ray induced  $^{14}\text{C}$  in the upper atmosphere produced from  $^{14}\text{N}$ , this has been demonstrated to be insignificant on a global-dose basis (Nair 1983): however, potential elevated local dose issues exist, and the option has so far not been embraced by waste authorities and regulators. Options for reducing the local impact have been investigated (Maceika *et al.* 2005).

More recently, with growing interest world-wide in carbon capture and storage from a range of industrial sources of carbon dioxide, the option to integrate  $^{14}\text{CO}_2$  from incineration of irradiated graphite wastes with other larger sources would result in a gaseous waste stream whose specific activity for  $^{14}\text{C}$  remains below the natural background. The isotopically mixed  $\text{CO}_2$  could be sequestered in porous rocks (former gas fields) (this is the strategy of ‘dilute and contain’; Wickham and Bradbury 2025). Gas re-injection has had a surge of interest from some companies and governments in the context of control of atmospheric carbon dioxide levels and releases unrelated to nuclear graphite oxidation (Stephenson *et al.*, 2019, Evans *et al.*, 2009). The possibility of incineration of irradiated graphite could be influenced by the growth (or non-growth) of such carbon dioxide gas re-injection technology deployments. Within the context of controlling atmospheric carbon dioxide levels, the UK government recently announced schemes to store carbon dioxide beneath the North Sea in December 2024 and, in liaison with the cement industry, under the Irish Sea (Department of Energy Security and Net Zero [DESNZ] UK 2024).

An integrated graphite-management process combining an alternative oxidation treatment with capture of the off-gases has been demonstrated on the pilot-plant scale by a consortium of UK organizations, in this case using plasma heating to oxidize the graphite. This is illustrated as an example (Figure 22) and commences with ‘nibble and vacuum’ graphite removal, followed by gasification through plasma heating and subsequent carbon dioxide sequestration (Theodosiou *et al.* 2018). The features of such a process may include:

- Pressure-swing absorption to concentrate isotopes such as  $^{14}\text{C}$  into a separate but much smaller waste stream has been investigated (Bradbury and Mason 2008) but relies on cryogenic temperatures with high power demand for its success.

- Deliberate wet leaching to remove activity has also been considered, although most work on leaching to date has been focused on the consequences of inadvertent leaching from waste repositories (Wickham and Bradbury 2008).
- Biological digestion of irradiated graphite has been considered, and it is understood that the former Pebble-Bed Modular Reactor (PBMR) company of South Africa, PBMR Ltd, claim to have patented a particular mix of bacteria which they consider would be efficient. There is also some evidence of such processes affecting the graphite ejected from Chernobyl Unit 4, where conversion to sugars occurred during the explosion which were then attacked by bacteria and fungal spores, which effectively made the ‘graphite’ partially water soluble (Wickham, 2007).

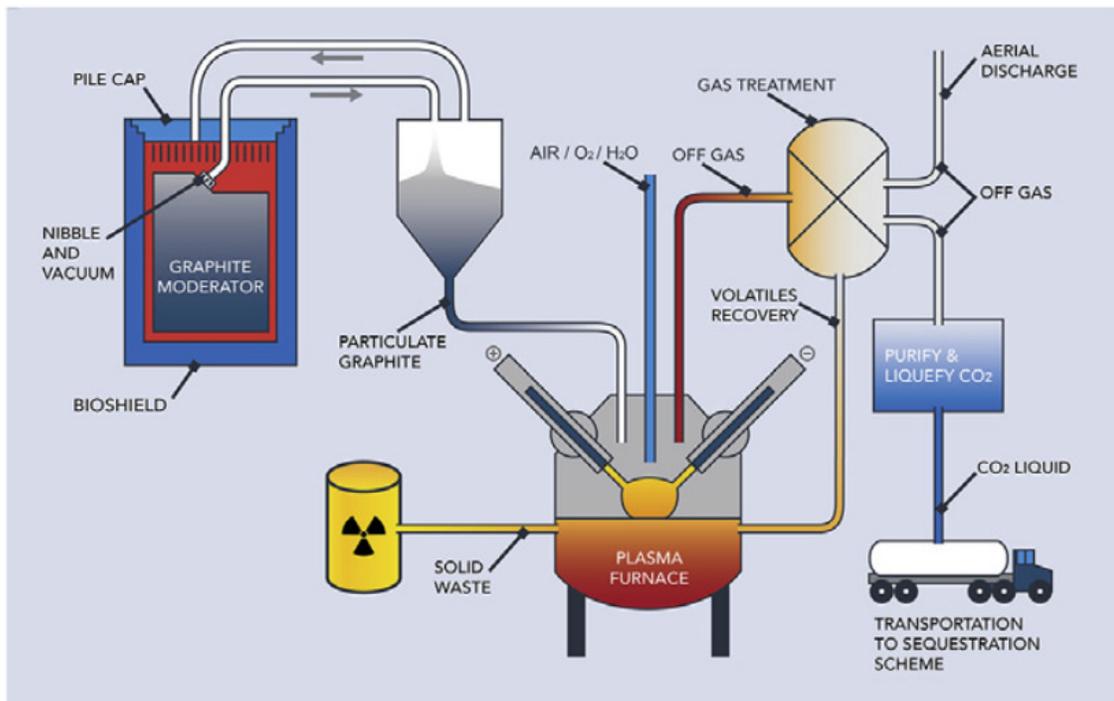


Figure 22. An illustrative ‘core-to-capture’ process for the removal and disposal of irradiated graphite, comprising ‘nibble and vacuum’, plasma oxidation, and sequestration of the carbon dioxide (Courtesy D. Bradbury, Sirius Analysis, and A. Theodosiou, The University of Manchester, UK)

- Electrical treatments. These are of two forms: electrical delamination of the graphite structure, as investigated by China during an IAEA CRP (Li and Wang 2016), and electrolysis in molten salt to extract fission products (Grebennikova *et al.* 2021, 2025).
- Vitrification. The benefits of vitrification are limited in application because there is a volume increase in the process (essentially reacting the graphite with titanium dioxide and aluminum – (Karlina *et al.* 2016)): however, it is under consideration by Russian specialists for stabilizing fission-product actinides from graphite which has become highly contaminated with debris from failed fuel elements.

- Recycling. The EU CARBOWASTE project (Grambow *et al.* 2013; Wareing *et al.* 2013) included investigations of recycling irradiated graphite into both new graphite components and other carbon-containing items for the nuclear industry such as activated-carbon filtration systems. An economic hurdle identified for industrial application was the need for manufacturers to have a separate production facility capable of handling radioactive material and re-manufacturing it into nuclear-grade graphite and doing so was not economically viable at the time of the CARBOWASTE project. Such recycling could become economically viable if the demand for recycling irradiate graphite were to increase significantly. There is a re-awakening of interest in such recycling with the advent of various designs of SMR. The first experiments for manufacturing new graphite from irradiated graphite were undertaken in the U.S. ‘Deep Burn’ project using irradiated high dose irradiated NBG-10 graphite material (Burchell and Pappano 2010). The approach was based on grinding graphite bent bars, mixing with pitch, and forming new “recycle” graphite. One complicating factor that had to be considered during the setting up of the recycle graphite fabrication line was minimization and containment of radioactive dust during grinding and forming. As a preliminary result, it could be stated that the ‘recycled’ unirradiated materials were comparable to the original quality. Recent studies (Liang *et al.* 2021) have identified that, after gasification to CO<sub>2</sub>, re-conversion to graphite can be conducted using lithium aluminum hydride at relatively low temperature: the product, akin to flakes of natural graphite, could then form the basis of new graphite products (as a filler in the conventional manufacturing route) or directly in the formation of new A3-type material for fuel pebbles or compacts and graphitic core components.
- Incorporation of irradiated graphite into the cement matrix stabilizing other forms of radioactive waste: this is ideal when the source of irradiated graphite is relatively small (as in the case of material from two experimental facilities at the Paul Scherrer Institute in Switzerland (IAEA 2023)) and could be applied for other small sources such as TRIGA research reactors, neutron-pulse devices, etc.
- Using the pore structure of graphite as a ‘repository’ for other hazardous waste in the form of a vitreous matrix.
- Use of supercritical carbon dioxide as a means of eluting radioisotopes (an option which is currently under investigation at the University of Mannheim in Germany).
- Chemical intercalation, leading to exfoliation, in order to release radioactive isotopes and remove them from the bulk of the material. This has been considered particularly in relation to material from the Chernobyl accident and the Italian Latina Magnox plant (Capone *et al.* 2019).
- Digestion and carbiding by hydrogen peroxide and molybdenum oxide to deal with heavily fuel-contaminated graphite (Pang *et al.* 2022): overall ‘digestion’ of the carbon matrix is also claimed to be possible.
- Minor effects. Here we include observations such as the reported release of <sup>36</sup>Cl during grinding of irradiated graphite.

This is by no means an exhaustive list, but it covers the main research streams and discussion items which have been part of recent international collaborations that the report authors are aware of. Some proposed graphite treatment options have not been widely peer reviewed and have not been demonstrated for suitability at industrial scales. For example, it has been proposed that so-called ‘Browns Gas’ will reduce radioactivity in general wastes (Bramhall 2023), but the authors are not aware of any peer-reviewed studies investigating such treatment and are not aware of any studies to demonstrate that it would be suitable for industrial scale.

For completeness we add one further initiative which is perhaps strictly more ‘storage’ or ‘disposal’ than treatment, but which may also suggest applications for movement of material during processing: Deep injection of water-borne pulverized graphite into suitable geological strata such as exhausted gas fields: such gas storage will have been in porous rocks and will have been contained by hydraulic pressure, which should therefore be a reversible option for suspended wastes. This process is analogous to ‘fracking’, which is subject to strict controls and in some countries is banned altogether.

Somewhat related to the previous example, Pavliuk, Kotlyarevsky *et al.* (2022) have investigated suitable treatments for graphite in aqueous sludges which have been encountered in various cavities and vaults in certain reactors, commencing with their characterization.

## 9 Graphitic Fuel Matrix Material

TRISO Coated Particles (CPs) are embedded in the A3-type matrix carbon. This has good thermal conductivity, and its unique carbonaceous structure allows for efficient heat transfer for dissipating the thermal energy generated by the coated fissile heavy metal. The graphitic matrix is also a neutron moderator. In addition, the matrix provides mechanical protection for the embedded fuel particles especially in case of spherical HTGR fuel elements when subjected to high mechanical forces during pneumatic transfer within the fuel handling system, including potential free fall on top of the reactor core and significant Hertzian contact pressures with neighboring fuel pebbles and with the reflector walls during the passage towards the core discharge channel.

The SiC layer of the TRISO particle starts to decompose at temperatures around 1800 °C with rapid degradation at >2000 °C (Wells *et al.* 2021, Zhou and Tang 2011). Therefore, the graphitic matrix for the TRISO fuel pebbles or compacts cannot be graphitized at the much higher temperatures and for durations typical for industrial graphite manufacture. They are heat-treated around 1950 °C for much shorter duration. The raw A3-3 material carbon mixture typically contains ~72% natural graphite<sup>20</sup>, ~18% synthetic graphite particles and ~10% binder (IAEA 2012).

The desirable characteristics for such a carbon matrix material have been identified as (Schulze *et al.* 1982):

- High carbon density
- High thermal conductivity
- High mechanical strength
- Low Young's modulus

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<sup>20</sup> Natural flake graphite is much less pure than synthetic nuclear graphite, and therefore the purification stage in A3-type material is extremely important to avoid reactivity losses.

- Good corrosion resistance
- Small thermal expansion
- Low anisotropy
- Good dimensional stability under high-flux irradiation with fast neutrons
- Lowest possible neutron activatable metal (impurity) content

In order to obtain the desired properties, the following manufacturing variables must be controlled:

- Sources of natural graphite,
- Sources of synthetic graphite,
- Grain size of filler particles including micro/nano-beads,
- Ratios of filler to binder contents,
- Types and content of binder resins without or with hardener additions,
- Heat treatment temperature and duration.

Such experimental variations resulted in the sequential numbering of the German graphitic matrices based on the A3-type and has been applied in TRISO fuel and HTGR developments in many countries.

As well as the inability to graphitize the binder in the presence of the fuel particles, since the SiC layer would be compromised, there is also no possibility for re-impregnation to gain a higher carbon density.

### 9.1 Manufacture of A3 Graphitic Fuel Matrices

The quality of TRISO fuel is determined by the structure of the coating layers and by the surrounding graphitic matrix. The manufacture of spherical fuel elements and cylindrical fuel compacts (used in the hexagonal block-type HTGR fuel elements) follow defined manufacture specifications, which have been applied to the U.S. AGR irradiation experiment series (Demkowicz *et al.* 2023).

The manufacture of spherical HTGR fuel (pebbles) begins with the grinding and mixing of the natural and synthetic graphite flakes with the binder resin, which is a thermoplastic phenol resin dissolved in methanol, in case of the A3-3 matrix type. An alternative is the A3-27 variant where the graphite flakes are mixed with phenol and hexamethylenetetramine at 130 °C. This results in more precise composition limits in the A3-27 compared with A3-3. The resin in the A3-27 case was not a precursor but was synthesized during the following heat treatment steps. The compositions of A3-3 and A3-27 after the final manufacture process are shown in Table 5 (from IAEA 2012). Table 5 defines the differences between A3-3 and A3-27.

Table 5: A3 matrix composition after final heat treatment (from IAEA 2012)

Component	A3-3	A3-27
Heat Treatment at	1800 or 1950 °C	1950 °C
Natural graphite [%]	72	71.2

Graphitized coke [%]	18	17.8
Binder [%]	10	11

The main difference between A3-3 and A3-27 is the use of a slightly higher synthesized binder resin content with and an enhanced heat treatment temperature, which has later on also been applied to the A3-3 matrix. A more favorable behavior under irradiation and corrosion conditions resulted from these slight deviations in the manufacturing procedures.

Figure 23 shows the principles of the production route for fuel pebbles:

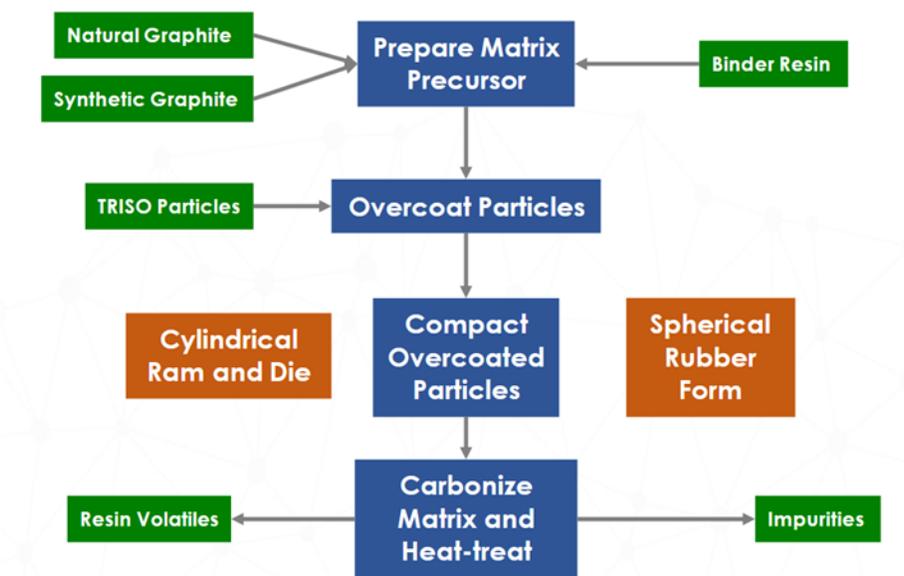


Figure 23: Manufacture process of TRISO fuel elements (from Demkowicz *et al.* 2019)

A part of the prepared matrix precursor is used for the over-coating of the TRISO fuel particles, which is an important step to minimize any potential damage due to direct contact of the TRISO CPs (Demkowicz *et al.* 2019). The over-coating has a thickness of  $\sim 200 \mu\text{m}$  and is controlled before adding the over-coated particles to the matrix mixture for forming either spherical or cylindrical compact fuel types.

In the case of the pebble type fuel, the process of isostatic molding is used to achieve as near isotropy as possible. This ambient temperature process involves using silicon rubber dies at an applied pressure of about 3 MPa for the pre-molding of the spherical fuel zone (see Figure 24, top left). The fuel-free outer shell is surrounded by the rubber dies during the final molding at 300 MPa (Hackstein *et al.* 1985). These high pressures facilitate the densification without the need for subsequent impregnation steps. The significance of the proper overcoat for the TRISO CPs is crucial for this high-pressure processing step and for the irradiation behavior of the TRISO fuel. The graphitic matrix of the pebble fuel also acts as a neutron moderator and needs a high carbon density. Mechanical lathing is used to obtain the diameter of 60 mm. The total heavy metal content (i.e. the maximum ‘density’ of TRISO

CPs per pebble) is controlled, because a higher CP-content lowers the mechanical strength of the pebble and consequently increases the failure rate of the TRISO particles during irradiation.

The manufacture of the cylindrical fuel compacts evolved over the past decades and did not initially have over-coatings of the TRISO CPs (Bresnik 1991). For the FSV fuel compact manufacture, a 40-cavity molding device was filled with CPs, and the matrix mixture was then injected into the heated molds. The baked compacts were ejected after cooling and have successively undergone thermal carbonizing, HCl leaching and high temperature heat treatment steps.

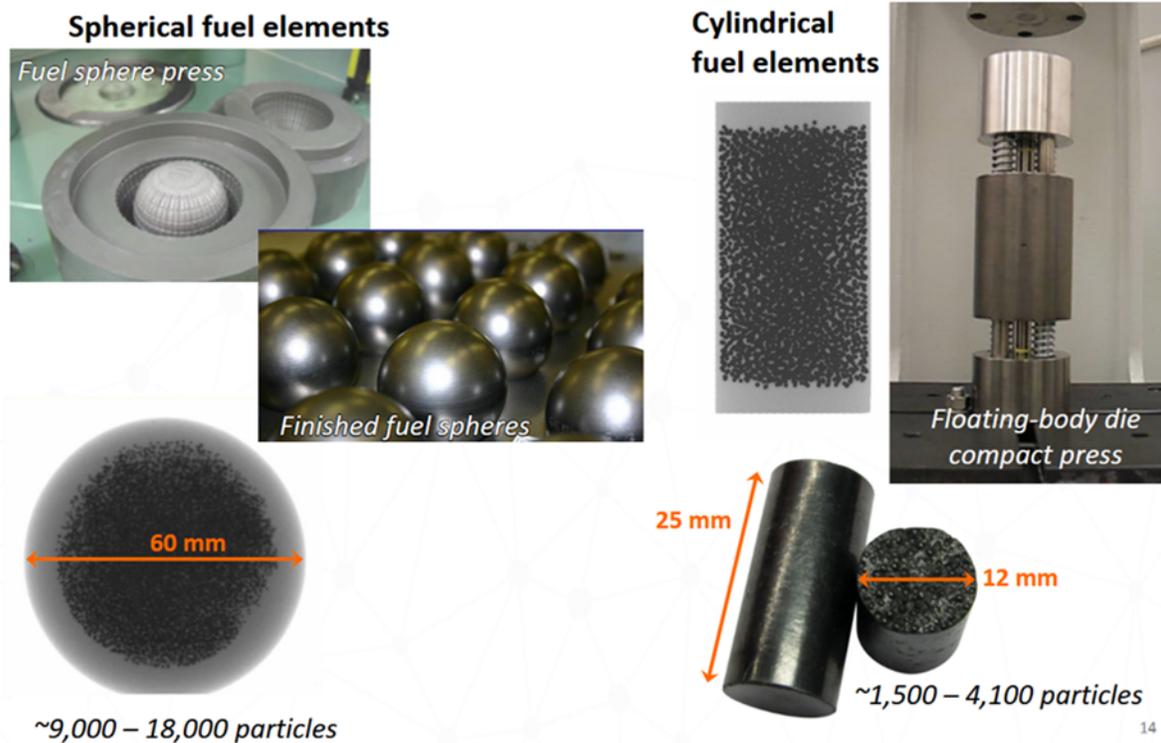


Figure 24: Manufacture steps for spherical and cylindric HTGR fuel (from Demkowicz et al. 2019)

Figure 25 shows a significant reduction of defects in the SiC layer after the adaptation of the over-coating technology for the HTGR compact manufacture in the U.S.

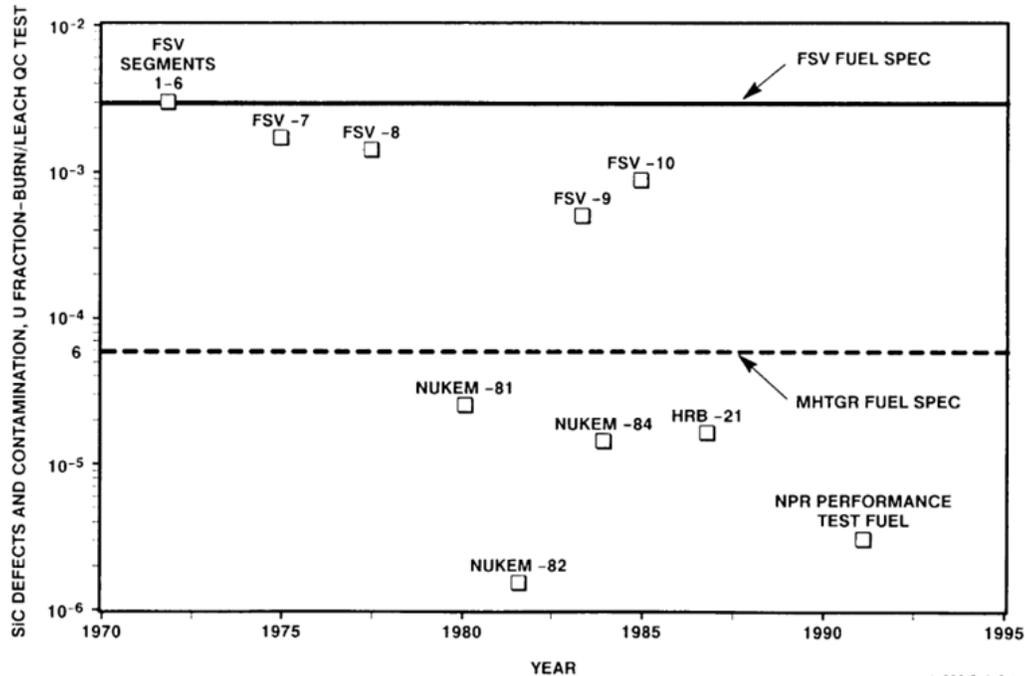


Figure 25: Evolution of HTGR fuel compact quality in U.S. (from Bresnick 1999)

Uniaxial compaction provides the final shape and dimensions required without a need for mechanical machining. The density of the CPs in this type of fuel is significantly higher than that in the pebble fuel. Between the AGR-1 and the AGR-2 irradiation campaigns, the compacting process was further improved by enhancing the temperature and pressure of compaction. Also, warm pressing instead of methanol soaked overcoated particles and a change from simple die to double-acting floating body press produced higher quality fuel (Phillips 2010).

Such improvement in the manufacturing process allowed higher power densities for the block-type fueled cores associated with extremely low fuel particle failures during the AGR program (Demkovicz *et al.* 2017).

The over-coating process in Germany was done in a rotating drum filled with CPs and the carbon-matrix mixture. The quality of the over-coating was rather dependent upon the manual operation. This process has also been improved to be done in a reproducible automated way.

As is the case for nuclear graphite, the Heavy Metal Concentration (HMC) has to be controlled also in the carbon matrix and in the outer PyC layer material because fission products from residual dispersed uranium in the matrix and in the outer PyC layer will not be retained.

## 9.2 Features of the Graphitic Fuel Matrix Material

It is useful to consider the manufacture and properties of the graphitic matrix material in a little more detail. The pre-formed ‘body’ of the graphitic matrix is carbonized at about 800 °C in an inert gas atmosphere and undergoes a short (~1 hour) high-temperature treatment at maximal 1950 °C under vacuum for degassing and purification. The binder is not graphitized (as discussed earlier) but only yields a ‘glassy carbon’ (GC) or ‘glass-like carbon’ structure, also known as ‘vitreous carbon’. GCs are obtained by the pyrolysis of carbon-rich precursor polymers in an inert atmosphere of, typically, Ar or N<sub>2</sub> and only occasionally in vacuum (Uskoković 2021).

The GC is a non-graphitized, or non-graphitizable, carbon, which combines glassy and ceramic properties with those of graphite. This material has a fullerene-like microstructure and exhibits high thermal stability, high thermal conductivity, hardness (5-6 Mohs), low density, moderate electrical conductivity, low friction, high resistance to chemical attack, and high impermeability to gases and liquids (Cappelletti *et al.* 2018).

Studies on the pore structure within A3-3 material showed that the volatilization of binder during the high-temperature treatment at >1800 °C led to a porosity of 17%; but the density of the carbonized skeleton improved (Zhou *et al.* 2018). The micropore structure in the graphitic fuel matrix seems to determine the materials properties. It is influenced by the molding pressure, producing mainly closed pores, and the escape of gases during carbonization process leaving open porosity behind (Zhang *et al.* 2019).

The micropore structure of the A3 matrix influences leaching, corrosion, and oxidation effects but may also influence the access, adsorption and activation of impurities such as nitrogen, as a precursor for <sup>14</sup>C.

The <sup>14</sup>C profile in an irradiated AVR moderator pebble (without fuel particles) has been measured after about 1500 Full Power Days (Nieder and Straeter 1988). As can be seen in Figure 26, there is a strong increase in <sup>14</sup>C activity at the pebble surface and a nearly constant activity with a small slope towards the center of the pebble. However, this <sup>14</sup>C activity level within the fuel pebble is about 4-5 times higher than the calculated value stemming from the neutron-activation of <sup>13</sup>C.

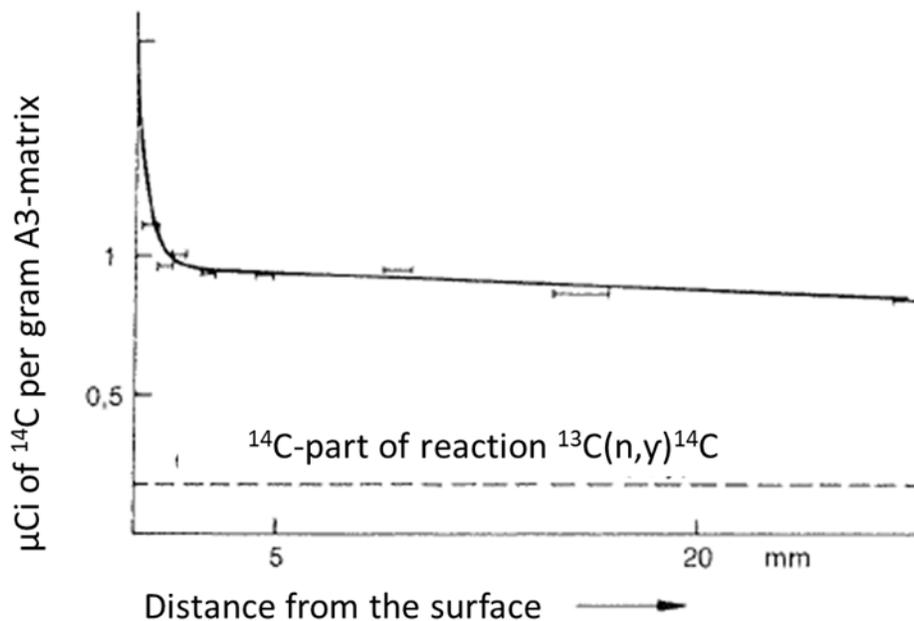


Figure 26: <sup>14</sup>C profile in an A3-matrix pebble after 1500 Full Power Days (from Nieder and Straeter 1988)

This means that a flat concentration of <sup>14</sup>N must already have existed within the pore structure of the A3-3 matrix material. An explanation could be the aforementioned higher fraction of open porosity compared to nuclear graphite.

A similar measurement has been undertaken for the distribution of tritium in an A3-3 pebble after a residence time of about 7 years within the AVR core (see Figure 27):

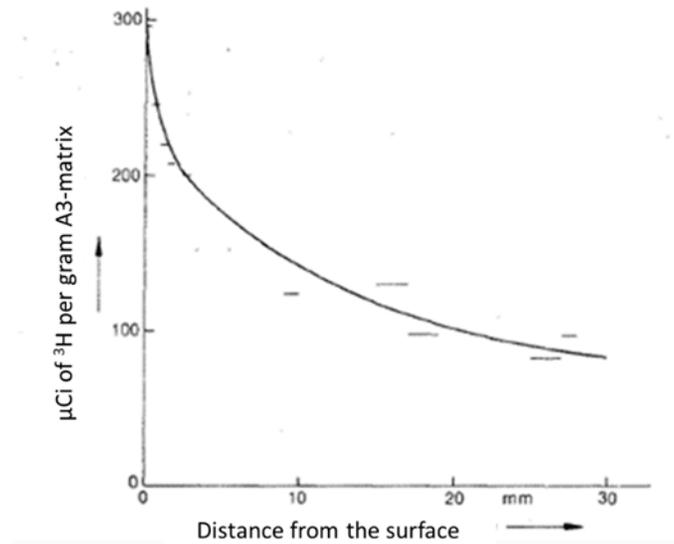


Figure 27: Tritium profile within an A3-matrix pebble (from Nieder and Straeter 1988)

Assuming an average value of 150  $\mu\text{Ci}$  per gram of A3-matrix equal to  $5.5 \times 10^6$  Bq/g, the authors postulated a hydrogen/tritium content within the pore structure of the A3-pebbles and of the graphite reflector of 1.6 kg. (Nieder and Straeter 1988). The tritium/hydrogen content in the coolant gas is judged to be only in the range of 1g. This means that hydrogen/tritium is ‘gettered’ (selectively accumulated) within the pore structure of the A3-matrices (and in the graphite structures).

The co-existence of hydrogen (and nitrogen) within the pore structures of the carbonaceous materials in an HTGR core might also explain the creation of organic functional groups *via* irradiation- and temperature-induced chemical reactions. Similar measurements have been made with A3-fuel pebbles (Wenzel *et al.* 1979) and within the graphite spines of the Peach Bottom Unit 1 spent fuel elements (Wichner 1980) – Figure 28:

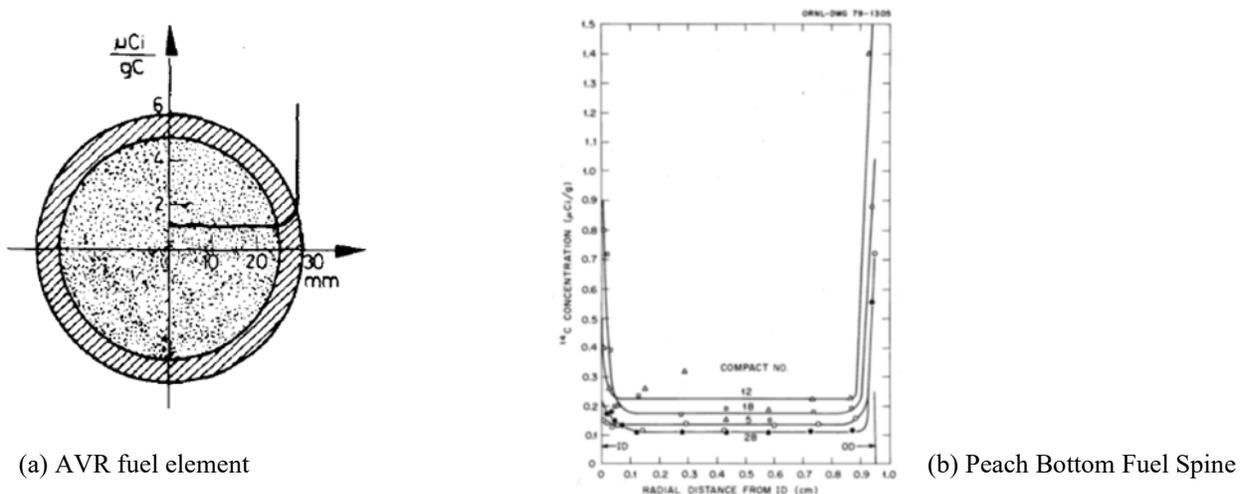


Figure 28:  $^{14}\text{C}$  activities in AVR (a, from Wenzel *et al.* 1979) and Peach Bottom (b, from Wichner 1980)

These effects and phenomena will also not only influence the release kinetics of radionuclides (like tritium, radiocarbon etc.) during transport, storage and disposal but also potential options for treatment and conditioning of this kind of radioactive waste.

The behavior of fission products has not been considered here. This would have to be done in another context addressing the fission-product retention capabilities of the TRISO fuel itself.

## 10 Separation of Spent TRISO Fuel from Fuel Elements

The possibility of separating spent TRISO fuel compacts from the surrounding graphite fuel block will not be dependent on the TRISO manufacture and coating, as long as there are no TRISO particles directly contacting the graphite, due to a fuel-free zone at the surface of the ‘modern’ TRISO fuel compacts. Decomposing spent fuel compacts or spent fuel pebbles is a challenge because the highly burned-up TRISO particles have experienced irradiation-induced property changes and possess extremely high internal pressure. This is indeed influenced by the manufacturer’s quality program.

### 10.1 Introduction & Overview

The operational waste volume issue related to spent fuel is primarily dependent on the low power density of helium-cooled HTGRs and to a more limited extent also to molten salt cooled AHTRs:

- LWR: 50 - 100 MW<sub>th</sub>/m<sup>3</sup> (IAEA 2020)
- SFR<sup>21</sup>: ~300 MW<sub>th</sub>/m<sup>3</sup> (EdF 2022)
- HTGR: 2 – 8 MW<sub>th</sub>/m<sup>3</sup> (von Lensa *et al.* 2020)
- AHTR: <20 MW<sub>th</sub>/m<sup>3</sup> (IAEA 2023)

The periodic exchange of irradiated reflector/moderator blocks will also generate waste to be stored and conditioned for waste disposal. This is also a significant difference e.g. to LWRs with a liquid moderator/reflector medium, where the regulated management of liquid wastes already has existing practices and standards (NRC 2025a). The same level of experience does not yet exist to establish practices and standards for the handling, conditioning and waste minimization of irradiated graphite and carbonaceous materials including e.g. particulate dust, adhesive soot and surface contamination.

The low power densities are mainly a consequence of the reduced heat transfer capabilities of gases compared to water, liquid metals and molten salt coolants. Inherent or passive safety approaches relying on ultimate heat transfer by heat conduction and radiation combined with suppressed natural coolant convection phenomena also result in lower power densities e.g. for modular HTGR concepts. The core geometry of modular HTGRs is deviating from the usual 1:1 diameter to height ratio for a neutronically optimized core configuration in favor of enhanced passive heat transfer *via* the core structure and reactor pressure vessel surfaces. This leads to higher neutron losses requiring slightly larger volumes of graphite core structures.

Figure 29 shows the different HTGR fuel elements with spherical and block-type fuel elements (Fütterer *et al.* 2014). In Japan, a ‘pin-in-block’ fuel element has been developed and tested, which places the fuel compacts into a hollow graphite rod (pin) to be inserted into a graphite block. The fuel

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<sup>21</sup> Sodium Fast Reactor

pins can easily be removed and stored or disposed of separately from the graphite block (Fukaya and Nishihara 2016).

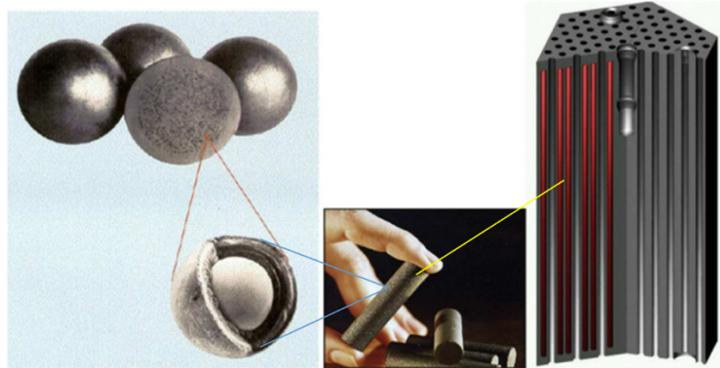


Figure 29: TRISO fuel in spherical and block-type HTGR fuel elements with fuel compacts (middle) (from Fütterer et al. 2014)

Whereas fuel elements from LWRs show a ~25% mass fraction of the fuel structures related to the initial heavy metal content (MIT 2011), HTGR fuel elements contain only 3-4 mass percent of initial heavy metal in the form of coated uranium oxide or uranium oxycarbide kernels. The rest is composed of PyC/SiC coatings, graphite or fuel carbon matrix. For waste management, the volume fractions are important and shown in the following figures for spherical (Verfondern and Nabielek 2017) and block-type HTGR elements (Figure 30).

96.6% of a fuel pebble volume is occupied by the A3 graphitic matrix and only 0.5% by the fuel kernels. In the case of block-type fuel, the volume of the graphite block (including bore holes for cooling and fuel compacts) takes 84%, the A3 carbon matrix of the fuel compacts about 10% and the fuel kernels about 1%. The PyC and SiC coatings represent 3-5 % of the fuel element volume, which is 5-6 times more than the volume of the initial heavy metal in the fuel kernels.

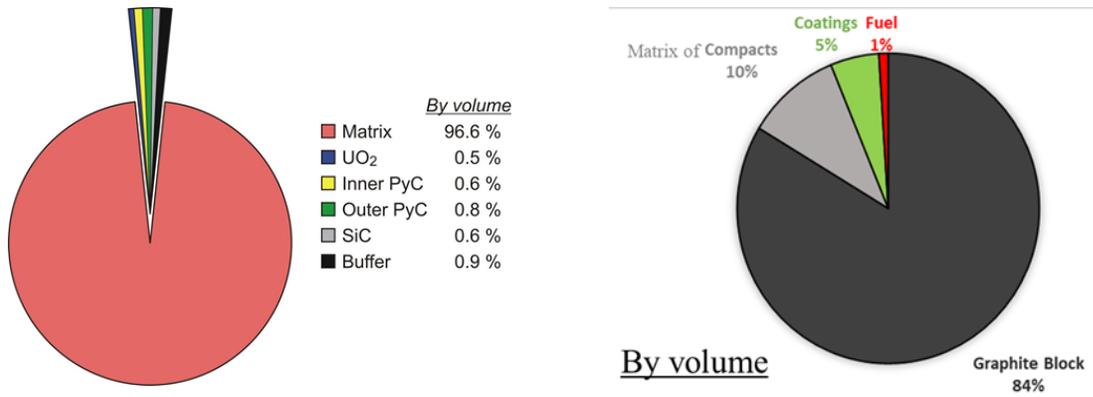


Figure 30: Volume fractions of HTGR fuel element components (spherical fuel on the left side (from Verfondern and Nabielek 2017), block-type on the right (from Forsberg 2024))

The consequences of these volume relationships with regards to HLW waste management are shown in Figure 31 (Kim 2024). The analysis leading to these figures was for the case that all advanced nuclear reactor designs will get equal share during the market introduction till 2050.

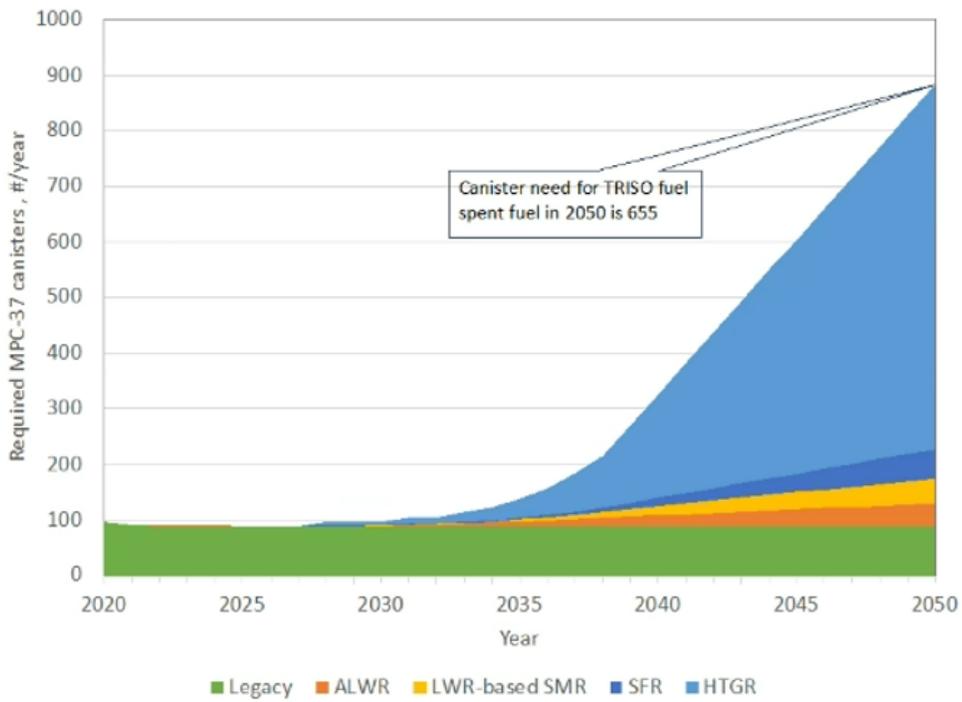
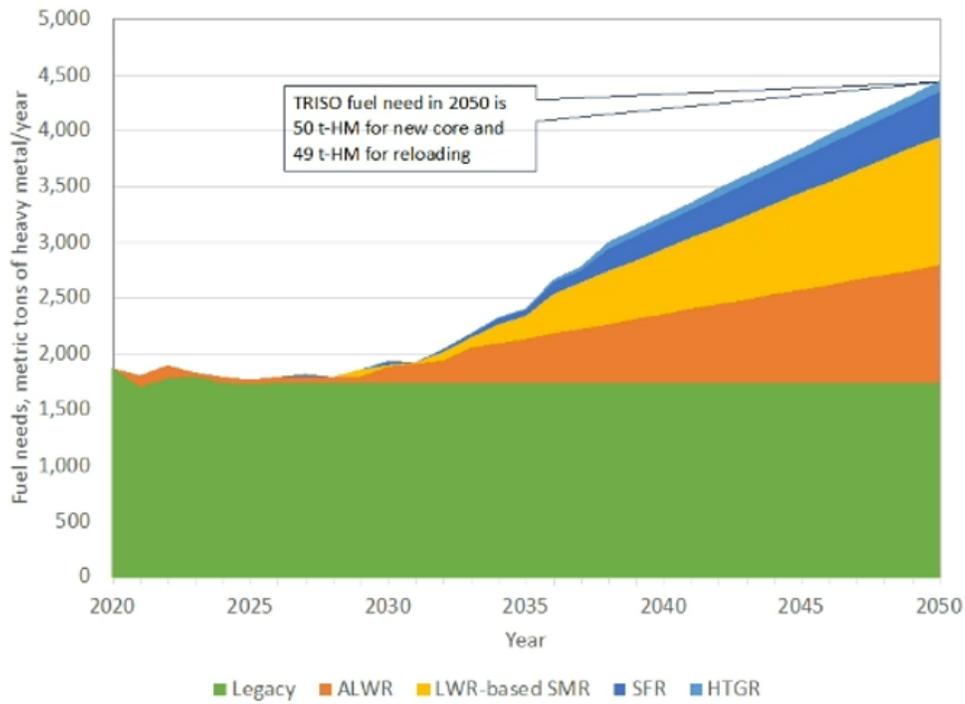


Figure 31: Initial fuel (upper image) and spent fuel canister needs (lower image) of HTGR relative to other reactor lines (from Kim 2024)

HTGRs have less need for fresh uranium fuel than earlier gas-cooled reactors due to higher thermal efficiencies and burn-up. However, the expected number of required spent fuel canisters for HTGRs is considerably larger compared to the needs of the other new and existing (legacy) nuclear reactors, *i.e.* 74% of the total canisters necessary for the spent fuel of the whole residual reactor fleet. These results are consistent with the above considerations on the volume relationships of fresh (and spent) fuel from LWRs and HTGRs.

Table 6 reflects the volume of the spent fuel for PWR, SFR and HTGR and other parameters that have an impact on the canister requirements. A Multi-Purpose Canister (MPC-37) was assumed for these comparisons.

Table 6: Spent fuel characteristics of PWR, SFR and HTGR (from Kim 2024)

	PWR w/ oxide	SFR w/ metal	HTGR w/ TRISO
Example reactor	Ap1000	Sodium	Xe-100
Required canisters, #/GWe-yr	1.1	1.5	20.3
SNF loading, assemblies or pebbles	37	37	39,006
Heavy metal loading, initial MT-U/canister	19.9	4.1	0.3
Decay heat @ 5 yrs, kW/canister	56.5	21.9	2.4
Decay heat @ 50 yrs, kW/canister	16.7	6.9	0.6
Decay heat @ 1,000 yrs, kW/canister	1.5	0.6	0.03
A2-values @ 5 yrs/canister	1.0E+07	2.8E+06	3.4E+05
A2-values @ 50 yrs/canister	8.4E+06	2.1E+06	2.2E+05
Gammas (>1 MeV) @ 5 yrs/canister	1.0E+16	2.3E+15	3.0E+14
Gammas (>1 MeV) @ 50 yrs/canister	2.2E+14	7.0E+13	6.0E+12
Fissile mass of fresh fuel, kg/canister	839	722	42
Fissile mass of discharged fuel, kg/canister	305	450	4

For HTGRs, the expected number of canisters per GWe/yr is nearly 20 times higher and the initial heavy metal loading is 66-times smaller than for LWRs. This also explains the much lower decay heat, the smaller A2 values for transportation and the lower gamma-radiation despite the higher burn-up of HTGR fuel. However, research may be needed to identify the potential benefits from these differences in terms of the canister's simplifications and/or benefits for storage, transport and disposal of spent HTGR fuel.

Volume reduction of the HTGR waste would be relatively straight-forward if similar canisters and waste management routines established for LWR could be applied to HTGRs. USA assessments (Richards 2002) have shown that the power density of HTGR fuel and the resulting low decay heat generation is much lower than in the LWR case. This allows for further volume reduction options within the limits of LWR canisters for spent fuel. Some of the waste volume reduction options are discussed further below. Different fuel element designs (spherical or block) may require specific approaches.

Many of the separation and volume reduction methods involve various physical methods for the disintegration of the A3-matrix, such as:

- Crushing, sieving, classification (mechanical separation by density etc.)
- Thermal shock treatment
- High-pressure water jet
- Ultrasonic treatment

- Electrical Pulsed-Power Fragmentation

Crushing and burning of fuel elements have mainly been developed in conjunction with the reprocessing of spent HTGR fuel. In some cases, the reprocessing of BISO or TRISO CPs involved damaging or breaking the particles to retrieve the uranium or thorium kernels. However, for most expected storage or disposal of separated TRISO fuel particles, including cases of waste volume reduction strategies, the coatings are usually envisioned to remain intact. Thus, methods which are known to damage the integrity of the TRISO particles were not considered further for this report.

## 10.2 Block-Type Fuel

Four HTGR types have already been built and operated, which are summarized under block type fuel elements in Table 7. The former Modular High-Temperature Gas-Cooled Reactor – Steam Cycle (MHTGR-SC) concept with 350 MWth is an example of a modular HTGR with block-type fuel elements. MHTGR-SC has undergone a safety analysis ('Pre-application Safety Evaluation Report for the Modular High-Temperature Reactor' (Williams *et al.* 1989)) and shares some design features with the NGNP, for which the pre-licensing interactions were suspended in 2013 (NRC 2025b).

The Peach Bottom Unit 1 reactor had a *de facto* 144-inch-long rod-type fuel element, which could be extracted from and reloaded into a core with permanent block-type graphite moderator elements. It is noteworthy that Peach Bottom Unit 1 and FSV contained uranium (and thorium) carbide kernels. If the coatings are not intact (e.g. in the first core of Peach Bottom Unit 1), the coatings will not provide a full contribution towards containment of the fissile material and fission products.

The HTTR design includes a 'Pin-in-Block' fuel element, which allows for an easy removal of the spent fuel pins, which contain fuel compacts within a thin graphite sleeve (pin). With the ability to remove the fuel pins and dispose of them separately from the rest of the fuel element, this fuel element allows a volume reduction of the HLW by about 80%, by design.

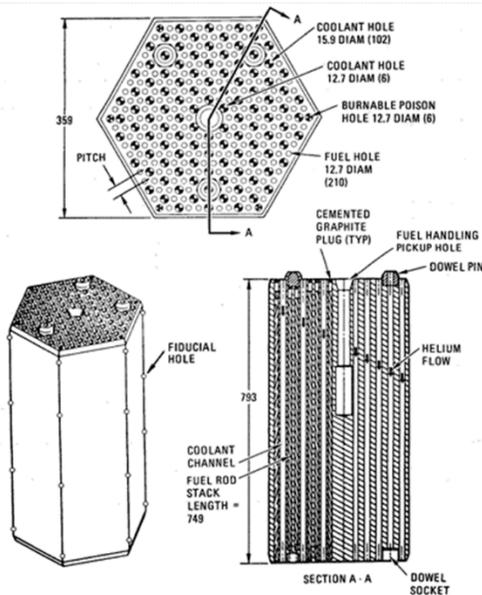
Systematic investigations on the graphite and fuel element structures after irradiation have only been sparsely reported and documented. However, such investigations are still possible in the context of decommissioning and waste conditioning (Marschman *et al.* 1993).

Table 7: Main data on block-type HTGRs (von Lensa *et al.* 2020 and IAEA 2022)

	Dragon	Peach Bottom	FSV	HTTR	MHTGR-SC
Thermal Power [MW]	21,5	115	842	30	350
Electric Power [MW]	-	40	330	-	150
Fuel / Core Type	Block	12 ft. Rods	Block	Pin-in-block	Block-Ring
Power Density [MW/m <sup>3</sup> ]	14	8,3	6,3	2,5	5,9
He Outlet Temperature [°C]	750	700-726	777	850-950	750
He Inlet Temperature [°C]	350	327	404	395	322
Flow Direction	upwards	upwards	downwards	downwards	downwards
Particle Type	BISO / TRISO	BISO	TRISO	TRISO	TRISO
Fuel Composition	(U, Th, Pu)O <sub>2</sub>	(U, Th)C <sub>2</sub>	(U,Th)C <sub>2</sub> & ThC <sub>2</sub>	UO <sub>2</sub>	UCO
Enrichment [wt%]	3.5 - 93	93	93	3-10	15,5
Operation Period	1964-1975	1966-1974	1976-1988	1998 ff	project
Years of Operation	11	8	12	27 ff	-
Decommissioning Status	in progress	Safestore	Site reuse	-	-

### 10.3 Mechanical Extraction

Irradiated fuel compacts have been removed from the FSV fuel elements No. 1-0743 and I-2415 (Saurwein *et al.* 1981). These fuel elements were pre-characterized before being loaded into the core. The PIE-activities were mainly restricted to physical irradiation-induced changes of the graphite and fuel compact structures. The removal of the spent fuel compacts from the graphite block was not foreseen in the design of the fuel elements. Thus, this procedure was provisionally developed for getting access to the irradiated fuel compacts (Figure 32).



#### Fuel element 1-0743 characteristics:

- Neutron dose:  $0.95 \times 10^{25} \text{ n/m}^2$
- H-327 graphite block
- 210 fuel holes with cemented graphite plugs on top
- 6 burnable poison holes
- 102 coolant holes
- In total 3130 fuel compacts

#### Fuel compacts:

- 12.5 mm (0.49 in.) in diameter
- 29.3 mm (1.94 in.) in length

#### Proposed Improvement:

- Conical plugs on bottom & top (Push-out)

Figure 32: FSV hexagonal block-type fuel element (from Vollman 2010) and fuel characteristics (from Saurwein *et al.* 1981)

For opening the fuel channels, a coring tool was developed, which removed the graphite at the bottom of the fuel channels and the cemented plugs on the top. The tool was fixed in the neighboring cooling channels to be exactly centered towards the fuel holes. Graphite scraps and dust were directly removed and collected. The push-out device was also fixed in neighboring coolant channels.

The fuel element 1-0743 contained different types of compacts:

- Compacts Cured In-Bed (CIB) during heat treatment in an alumina powder bed
- Compacts Cured In-Pile (CIP) during irradiation and operational temperature

The push-out forces were rather low ( $< 10 \text{ kPa}$ ) for a complete string of CIB fuel compacts within one fuel hole. The CIP compacts caused some problems, due to swelling during irradiation. However, the standard CIB compacts, which resemble modern fuel compacts, could easily be removed (Lord *et al.* 1993).

About 3% of the 3130 fuel compacts removed from the element were broken. Approximately two thirds of the broken compacts are thought to have been broken when pushed out of the block; the remaining one third of the broken compacts were probably broken prior to assembly of the element. Evidence of breakage prior to assembly was apparent in many instances (Marschman *et al.* 1993).

The irradiation-induced strains in the TRISO-particle fuel compacts were found to be small and somewhat anisotropic, with the axial strain exceeding the radial. It is observed that the predicted strains are about three times the measured strains (Figure 33). In addition, radial strains have been predicted to be greater than axial strains, but the opposite occurs. One possible explanation is that the model was developed primarily from design data in the fast fluence range 4 to  $10 \times 10^{25}$  n/m<sup>2</sup> and extrapolated to low fluence (Saurwein *et al.* 1981).

The difference between the predicted strain curves and the measured values could also potentially be partially due to creep effects due to the mechanical interaction of fuel compacts and the surrounding graphite fuel element structure.

For accelerating the removal of the compacts, a push-out device was developed, which allowed a synchronous removal of compacts from six fuel channels around a coolant hole. The prototype of the push-out device used a simple threaded-rod mechanism driven by an electric motor. The target was to remove all compacts from two spent fuel blocks within a shift of two operators. A technical layout of such mechanical volume reduction operations has been detailed by General Atomics and evaluated with the result that it is likely to be the only option which can be integrated into operation at a reactor site (Zimmerman 1984). It also generates little secondary waste, especially if the fuel compacts do not contain CPs directly exposed to the surface. However, coated-particle damage could still happen during push-out of fuel compacts being manufactured with the FSV fuel rod compacting technology. Figure 34 shows a sample compact, as manufactured at Idaho National Laboratories, with a protective layer on the peripheral CPs (Stempien *et al.* 2023).

Such push-out devices could be more automated, especially if the coring process could be simplified. This could potentially be achieved if removable conical or screwed plugs will be used at the bottom and at the top of the prismatic fuel block. The block design may be designed in a way that the fuel compact strings could as easily be pushed-out as it is done for the Pin-In-Block fuel elements.

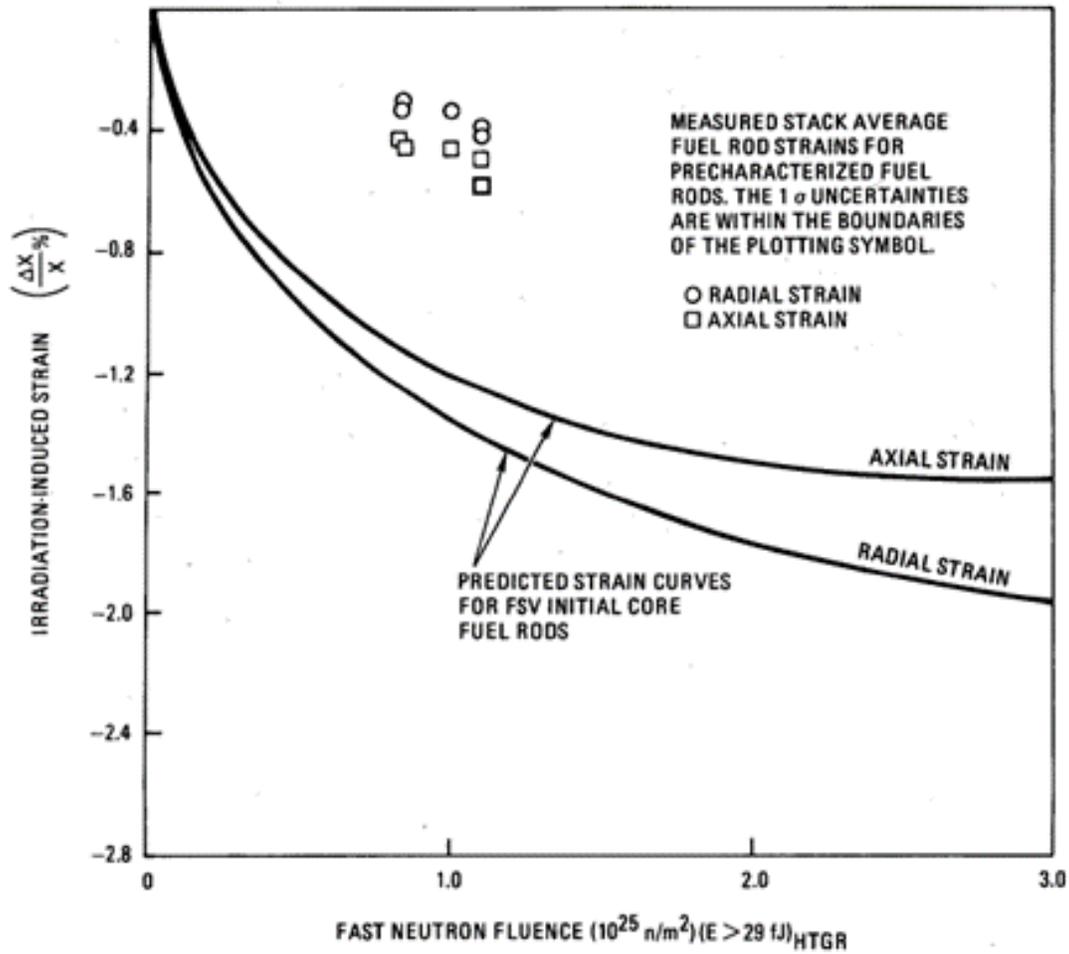


Figure 33: Comparison of calculated and measured strain for fuel compacts (from Saurwein et al. 1981)

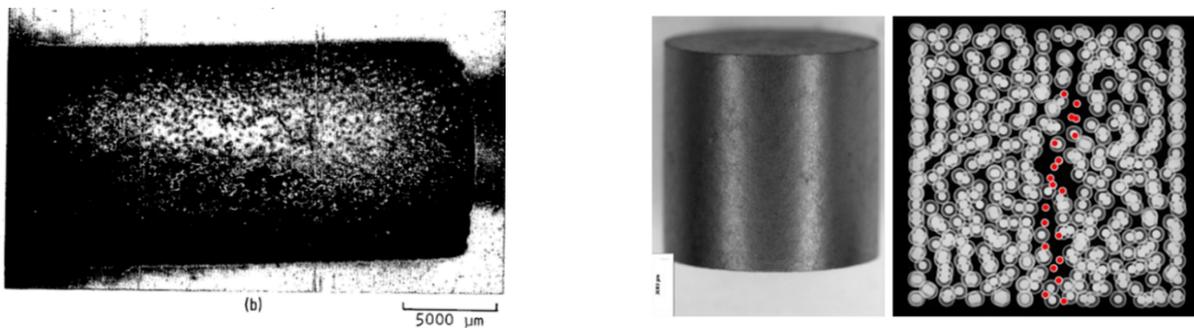


Figure 34: Extracted FSV fuel compact (left, Saurwein et al. 1981) vs. actual compact sample for irradiation tests (Stempien et al 2023)

#### 10.4 Pebble and Compact Disintegration

The TRISO particles in the spherical fuel elements and in the fuel compacts of block-type HTGRs are usually embedded in an A3-graphitic matrix. The disintegration methods for spent fuel pebbles may be applicable to spent fuel compacts (and *vice versa*).

Table 8 shows the main data of the HTGRs operated with spherical fuel elements and includes the U.S. Xe-100 HTGR project (Mulder *et al.* 2018), which is undergoing a design certification review from regulatory authorities in US and Canada.

Table 8: Main data on HTGRs with spherical fuel elements (from von Lensa *et al.* 2020 and Mulder 2018)

	<b>AVR</b>	<b>THTR</b>	<b>HTR-10</b>	<b>HTR-PM</b>	<b>Xe-100</b>
Thermal Power [MW]	46	750	10	250	200
Electric Power [MW]	15	296	2,5	105	80
Fuel / Core Type	Pebble	Pebble	Pebble	Pebble	Pebble
Power Density [MW/m <sup>3</sup> ]	2,6	6	2	3,22	4.8
He Outlet Temperature [°C]	850-950	750	700	750	750
He Inlet Temperature [°C]	275	250	250	250	268
Flow Direction	upwards	downwards	downwards	downwards	downwards
Particle Type	BISO / TRISO	BISO	TRISO	TRISO	TRISO
Fuel Composition	(U, Th)O <sub>2</sub>	(U, Th)O <sub>2</sub>	UO <sub>2</sub>	UO <sub>2</sub>	UCO
Enrichment [wt%]	LEU & HEU	93	17	8,5	15,5
Reactor Building	Containment	Confinement	Confinement	Confinement	Confinement
Operation Period	1967-1988	1985 -1991	2000 ff	2022	project
Years of Operation	21	6	24	2	-
Decommissioning Status	RPV Store	Safestore	-	-	-

The German AVR reactor was the prototype for the HTGRs with a pebble bed core being operated over 21 years. It was mainly used for mass tests of fuel elements with different kernels or coatings and for various safety tests. The Chinese HTR-10 was the first modular test-reactor and was followed by the 2 x 250 MW HTR-PM demonstration reactors, now under full power operation. China is also active in the development of waste volume reduction techniques for spherical fuel elements.

Physical and chemical disintegration methods will be presented, which have already been tested on a laboratory scale level or in the context of pilot-plants for reprocessing or recovery of fuel kernels from fuel manufacture charges not fulfilling the specifications. A part of this methodology has also been evaluated with regards to their technology readiness levels (Arm *et al.* 2022).

## 10.5 Thermal Shock Treatment

Within the European CARBOWASTE project, thermal shock exposure was investigated on fuel compacts containing TRISO particles with ZrO<sub>2</sub> dummy kernels (Guittonneau 2010). Thermal shocks are often induced by using lasers, electric arcs, plasmas or electrons beams to initiate quick temperature transients from room temperature to a higher temperature (Guittonneau *et al.* 2008). For quenching, the heated material was immersed in liquid nitrogen or cold water.

The A3 matrix contains ~90% graphitic filler particles and has a high thermal conductivity (~100 W m<sup>-1</sup> K<sup>-1</sup>) and a low thermal expansion coefficient (~5 x 10<sup>-6</sup> K<sup>-1</sup>). These properties are favorable for resistance to thermal shock.

A (non-irradiated) compact containing 10 % of TRISO particles was placed in a furnace for 15 min at 777K. At this temperature the oxidation under air was still tolerable. The hot fuel compact was

repeatedly transferred into a Dewar filled with liquid nitrogen (77K). But no visible damage was observed despite several transients with  $\Delta T = \sim 700\text{K}$ .

A second set of experiments was operating inversely from cold environment (liquid nitrogen) to warm environment (water at about 360K). The compacts were immersed in the liquid nitrogen for several minutes and then put into the hot water. Only a few cycles were required to breach the compacts into diverse fragments (see Figure 35).

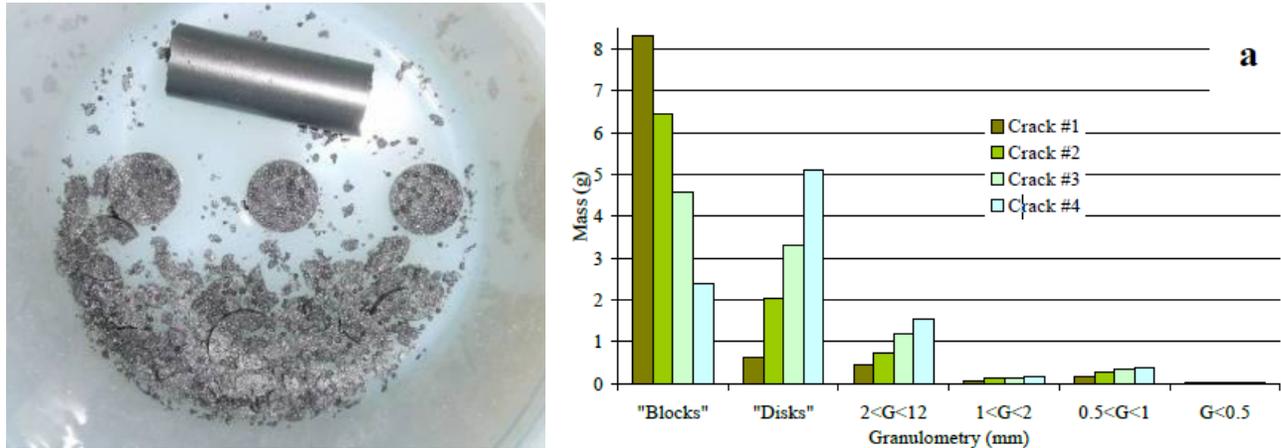


Figure 35: Defragmentation of dummy compact with 10% TRISO particles (from Guittoneau 2010, courtesy SUBATECH, France)

Retrieval of the TRISO particles was not satisfactory because the compacts disintegrated into little blocks, discs and granulate of different sizes.

The relatively small temperature transient of  $\sim 280\text{K}$  cannot explain the observed defragmentation. It is assumed that the liquid nitrogen enters into the open pore system of the A3-matrix and suddenly evaporates when submerged into hot water. This generates a high pressure within the pores and disintegrates the compacts.

The methods might be applicable as a pre-treatment for segmenting the fuel compacts into smaller fragments but not for separating all TRISO particles.

## 10.6 High-Pressure Water Jet Treatment

Another straight-forward method was the application of high-pressure water jets, which are applied for different technical purposes (*e.g.* cutting diverse materials including concrete, ceramics, wood etc.). Also, abrasive particles can be added into the high-pressure jet.

In published work, dummy fuel compacts were put into a cage with 14 mm diameter and a 500  $\mu\text{m}$  mesh size, which contained the ( $\sim 900 \mu\text{m}$ ) TRISO particles while releasing loose graphite particles. (Guittoneau 2010). Figure 36 shows the experimental arrangement, which could provide a pressure up to 4000 bars and moved the water jet in a pre-determined scheme and orientation over the cage.

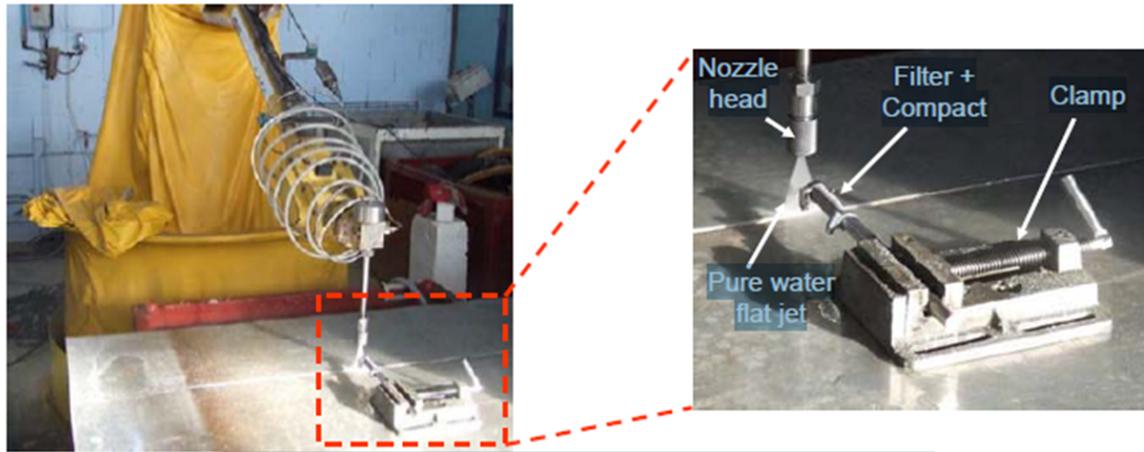


Figure 36: Experimental arrangement for water jet treatment (from Guittonneau 2010, courtesy SUBATECH France)

As no experience on the erosion of graphite was available, the pressure was varied between 500 to 1670 bars with different orientations of the nozzle relative to the compact. The results are shown in Table 9:

Table 9: Overview of high-pressure tests on fuel compacts (from Guittonneau 2010, courtesy SUBATECH France)

Test #	Jet orientation*	TRISO Vol%	Total TRISO	Intact TRISO	Damaged TRISO**	TRISO without coating***	Effective duration of the test	Lost graphite	Graphite erosion rate (%/min)
A	Perpendicular	9.9	400	394 (98.5%)	6 (1.5%)	0 (0%)	120 s	31.1%	15.6
B	Parallel	9.9	521	237 (45.5%)	48 (9.2%)	236 (45.3%)	165 s	50.1%	18.2
C	Perpendicular	20.9	1662	1425 (85.7%)	140 (8.4%)	97 (5.8%)	114 s	69.3%	36.5

(\*) perpendicular orientation indicates that the jet is perpendicular to the filter/compact system but its displacement is parallel to it.

(\*\*) oPyC layer is cracked or missing.

(\*\*\*) only  $ZrO_2$  kernel is still present.

Test A did not contain any heavily damaged TRISO particles (without coating) and only showed a few which have lost their outer PyC layer. The sorted particles are clean and free of the graphitic compact matrix.

In test B, the parameters are not adequate because a large fraction of the TRISO particles being broken, as seen in Figure 37, with 'residues' and sorted particles. Each black sphere is a  $ZrO_2$  kernel of 500  $\mu m$  diameter.

Results from test C appear promising. Erosion of one compact (36.5 %/min) corresponds to a graphite mass loss of about 0.12 kg/h. The 6 % fraction of fractured TRISO is certainly due to the non-optimized distance between the jet and the filter and/or the high TRISO packing fraction <20%. It is possible that the TRISO particles collided with each other and against the enclosing filter at very high speed, resulting in their coating failure.

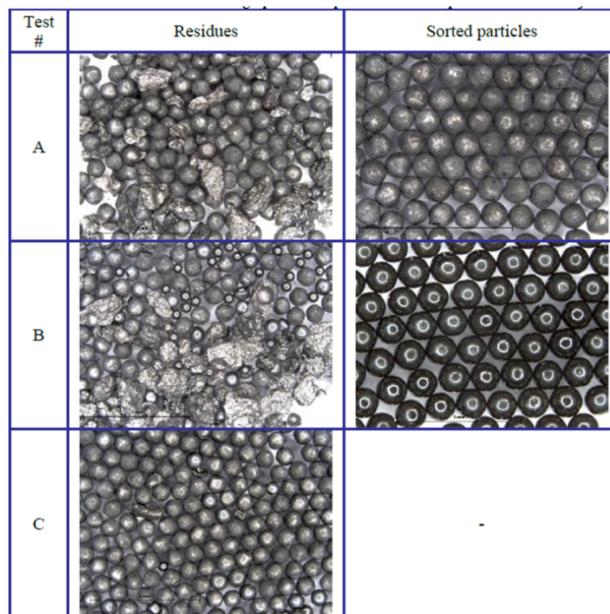


Figure 37: Photographs of residua and sorted particles (from Guittonneau 2010, courtesy SUBATECH France)

Thus, the water jet was able to erode graphite at a pressure starting at 500 bar but the optimal pressure to get a reasonable yield seems to be between 1000 and 1500 bar. The distance between the nozzle and the compact is a key parameter and must range between 20 and 30 mm and must be optimized as a function of the pressure and the angle of the jet. For this geometrical system, the displacement of the jet must be parallel to the compact with an orientation perpendicular to it, with cyclic displacement of about two seconds. The above tests demonstrate the proof of principle of the high-pressure jet treatment with water or other cryogenic fluids. For irradiated compacts (or pebbles), secondary waste is generated by the pressurized medium and the released contaminated matrix particles. Graphite and carbonized binder particles will be considerably contaminated in irradiated-fuel treatment. Contamination could be carried by the reflected high-velocity water spray into the surrounding area and will have to be collected and conditioned.

## 10.7 Ultrasonic Treatment

Ultrasonic fragmentation is based on cavitation effects and the implosion of bubbles in a liquid medium. Ultrasonic waves, which invade the solid matter, can also lead to fatigue effects supporting the disintegration of the samples. The energy intensity and the frequency range of the generator of the ultrasonic waves are the most important parameters. A dummy compact with 10% TRISO particle fuel has been exposed under water with four 35 kHz piezoelectric transducers of 80W each (Figure 38, Guittonneau 2010). The influence of diverse parameters on the fragmentation efficiencies has also been captured and discussed.

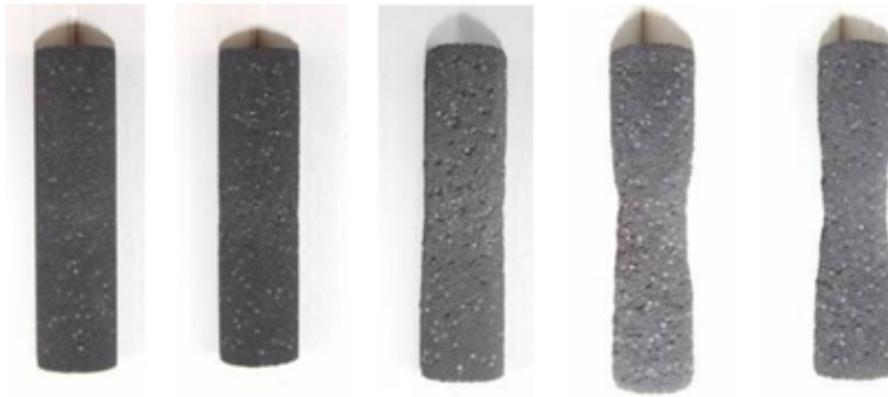


Figure 38: Eroded compacts after 2, 4, 9, 15 and 17.5 hours (from Guittonneau 2010, courtesy SUBBATECH France)

Figure 39 shows intact TRISO particles and a fractured piece of the outer PyC layer extracted with this technology.

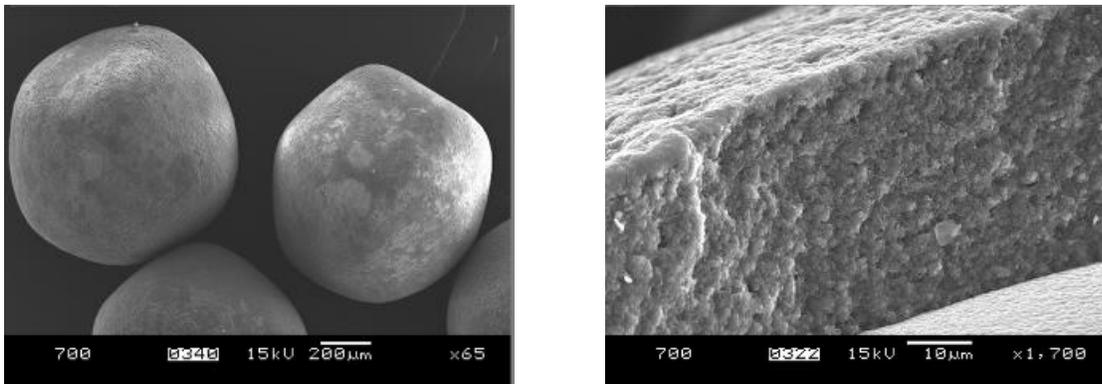


Figure 39: Extracted intact TRISO particle (left) and fractured piece of the outer PyC layer (from Guittonneau 2010, courtesy SUBBATECH France)

At higher power, the formation of new materials was observed, such as fullerenes  $C_{60}$ , nanotubes, fibers and rhombus-shaped pieces. High-energy ultrasonic waves also induce chemical reactions, as shown in Figure 40.

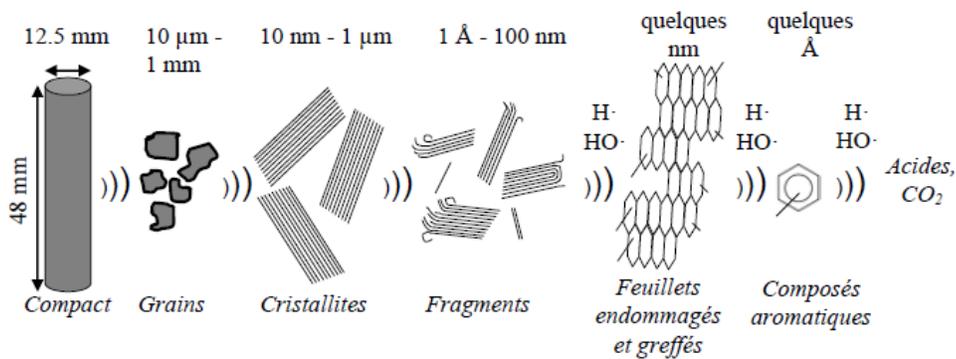


Figure 40: Scheme of degradation stages and induced chemical reactions (from Guittonneau 2010, courtesy DSUBATECH France)

The TRISO particles or kernels have been separated by flotation in bromoform ( $\text{CHBr}_3$ ). Fragmentation of fuel compacts and pebbles is energy intensive and leads to a large fraction of broken TRISO particles and is accompanied by induced complex chemical reactions. Thus, this technology may be more suitable for disintegrating TRISO particles than for extracting intact TRISO particles.

### 10.8 High-Voltage Pulse Fragmentation

Rock fragmentation is broadly applied e.g. in the mining industry. High-Voltage Pulse Fragmentation (HVPF) has become an alternative to mechanical milling, grinding and blasting technologies (Dakik *et al.* 2023). The high-voltage discharges in the order of several tens of nanoseconds in water create cavitation shockwaves, which allow a separation of diverse materials. This technology allows cleaning of surfaces, reduction of contamination and e.g. also liberation of inclusions, such as TRISO particles within an A3-matrix.

The principle of HVPF is illustrated in Figure 41.

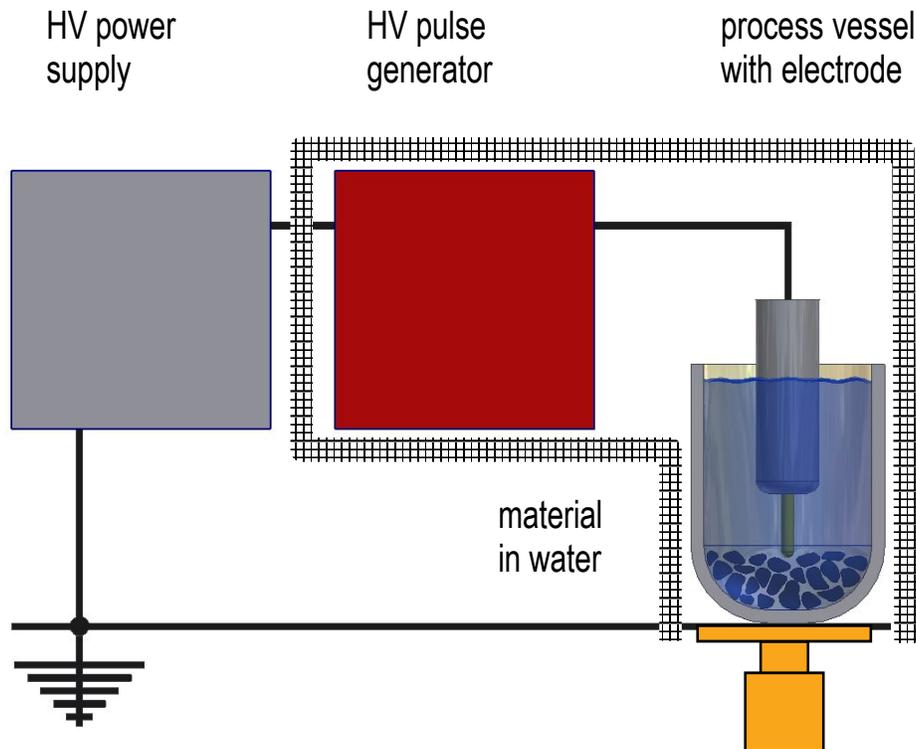


Figure 41: Principles of HVPF (from Fütterer *et al.* 2009)

At sufficiently short pulse rise times, the discharge preferentially crosses the material and disintegrates it along the grain boundaries. Important process parameters are the discharge voltage, pulse rise time, electric arc length, reaction volume, electric conductivity of the water and the number of discharges.

This technology was further adopted to the fragmentation of unirradiated dummy compacts with different TRISO particle densities (Masson *et al.* 2006). Surrogate spherical fuel elements were treated in a commercially available fragmentation machine (Figure 42, Fütterer *et al.* 2009).

The sample was fed into the water-filled reactor with a high-voltage electrode on the top and a grounded sieve on the bottom. The pulse generator repeatedly charged the electrode with voltages in the range of 200 – 400 kV, with maximum discharge currents about 10 kA.



Figure 42: Fragmentation installation with dummy of a spherical fuel element (from Fütterer et al. 2009)

After a few pulses the pebble was already crushed. After 1 minute of exposure (300 pulses) all CPs were liberated. Owing to their mechanical resistance, all CPs remained intact and could be separated by sieving (Figure 43).

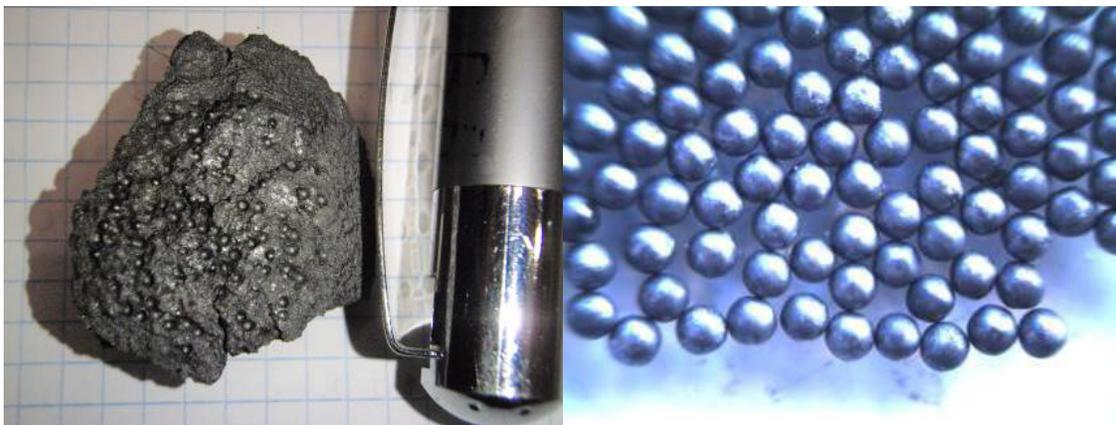


Figure 43: Crushed fuel pebble after 3 seconds (15 pulses) and liberated intact TRISO particles (from Fütterer et al. 2009)

During the treatment of fuel compacts some fractures of the TRISO particles were also observed. Releasing all particles from the fuel compacts, whilst not compromising the integrity of the TRISO particles, still needs further optimization of the parameters.

The experiments on non-irradiated surrogate fuel compacts and pebbles indicate the feasibility of the HVPF method. However, the tests need also to be conducted with irradiated samples to investigate

the influences of irradiation damage within the A3-matrix material and the TRISO coatings stressed by considerable internal gas pressure.

The electric energy for separation and fragmentation corresponds to a fraction of only between 0.125 and 0.25% of a pebble's lifetime electricity output.

### 10.9 Mechanical Fragmentation Options

With the exception of the extraction of complete fuel compacts from irradiated block-type graphite fuel elements, the physical fragmentation methods imply high mechanical forces on the A3-matrix and the TRISO particles, which challenges the integrity of the outer PyC layers or of the complete TRISO particles, especially if there is high internal fission gas pressure due to high burn-up. HVPF appears to be a potential option but has not yet been tested with irradiated fuel compacts or fuel pebbles.

The amount of solid secondary waste is limited to graphite dust and fragments of the A3-matrix material plus broken parts of the outer PyC layers. Liquid secondary waste arises for those fragmentation options in which a contact medium for transmitting mechanical forces is applied or using physical phenomena such as cavitation. Damaging the TRISO particle layers will generate HLW waste in the form of fragments of PyC and SiC layers as well as fractured TRISO particles, which do not fully retain the included fission products and provide access of media to the heavy metal kernel and the highly radioactive buffer layer.

Most mechanical fragmentation methods do not hinder further treatment and reuse of the irradiated graphite and the A3-matrix material.

Further tests with representative irradiated samples are necessary to validate the chosen options in a pilot scale facility. It is preferable, for such tests, to use simulant TRISO fuel e.g. with a boron carbide kernel, which does not generate high radiation levels (no significant fission products) but may provide information on the impact of representative internal helium gas pressures via the  $^{10}\text{B}(n, \alpha)^7\text{Li}$  reaction and consequences of irradiation on the mechanical stability of TRISO particles.

### 10.10 Chemical and Electrochemical Methods

Chemical and electrochemical methods for the separation of intact TRISO particles from the irradiated fuel compacts and fuel pebbles have been applied for Post-Irradiation Examination (PIE) within experimental HTGR fuel irradiation campaigns. Thus, prior experience exists on the deconsolidation of irradiated fuel compacts and fuel pebbles. An additional PIE requirement was the ability to identify also the local provenience of the extracted CPs within the fuel compact or of their radial position in a fuel pebble. However, this is not necessary within the framework of volume reduction strategies.

The chemical and electrochemical fragmentation options for fuel compacts and fuel pebbles likely benefit because the A3-matrix is not fully graphitized and contains a different pore structure with more open pores compared to graphite (Zhang *et al.* 2019). Sometimes fragmentation of carbonaceous materials by intercalation is considered as a physical method because the graphite crystal layers are expanded while they disintegrate. It is reported here under chemical and electrochemical methods as no direct physical forces are applied.

## 10.11 Homogeneous Oxidation

The oxidation of graphite and porous carbonaceous materials is divided into three domains:

- I. Chemical regime with homogeneous oxidation inside the whole volume
- II. Pore diffusion of oxidants with a corrosion profile at the surface of the material
- III. Mass transfer-controlled regime with preferred surface corrosion

A fourth regime, where the rate of oxidation is so high that the product is dominated by carbon monoxide rather than carbon dioxide, is sometimes recognized.

Homogeneous oxidation is accompanied by a high loss of mechanical strength. The oxidation rate in the first regime is lower as compared to the oxidation at higher temperatures. However, this can be overcome by applying specific catalysts, as shown in Figure 44:

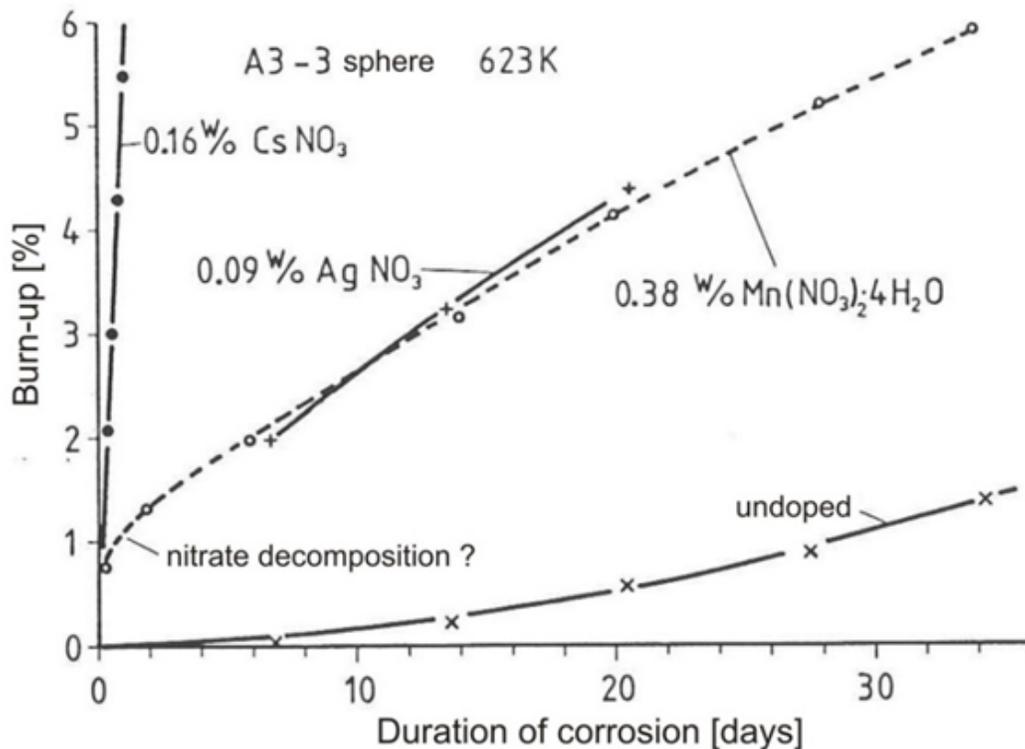


Figure 44: Homogeneous corrosion with and without catalysts (from Hinssen *et al.* 1982)

It can be seen in the leftmost trace that the addition of only 0.16% of  $\text{CsNO}_3$  strongly accelerates the oxidation of A3-matrix material at only 350-400°C (Hinssen *et al.* 1982) and disintegrates the A3-matrix within a few days (Figure 44).

This process can be further accelerated by removing the corroded bulk material progressively *e.g.* in a heated rotating drum and extracting the granular residua of the A3-matrix together with the loose TRISO particles. The released (unirradiated) TRISO particles essentially did not show any damage. Process-related damage to the TRISO particles can be practically ruled out. Optimum temperatures for the disintegration of un-doped spheres in air are between 820 and 870 K.

This process is validated on the laboratory scale but needs a scale-up to demonstrate co-processing of several pebbles (or compacts) in a batch or as a continuous process, in a semi-technical scale.

The secondary waste consists of solid granules originating from the graphite filler material, carbonaceous dust and <20% by weight of carbon dioxide, which also contains  $^{14}\text{C}$ . The carbon dioxide can be solidified *e.g.* as a carbonate, or as solid carbon to retain  $^{14}\text{C}$ . The carbon granulate can be mixed with a binder and pressed into a form, which can be disposed with or without heat treatment <1200°C or reused as an additive to concrete for enhancing the heat conductivity *e.g.* of concrete waste containers or backfill (IAEA 2016).

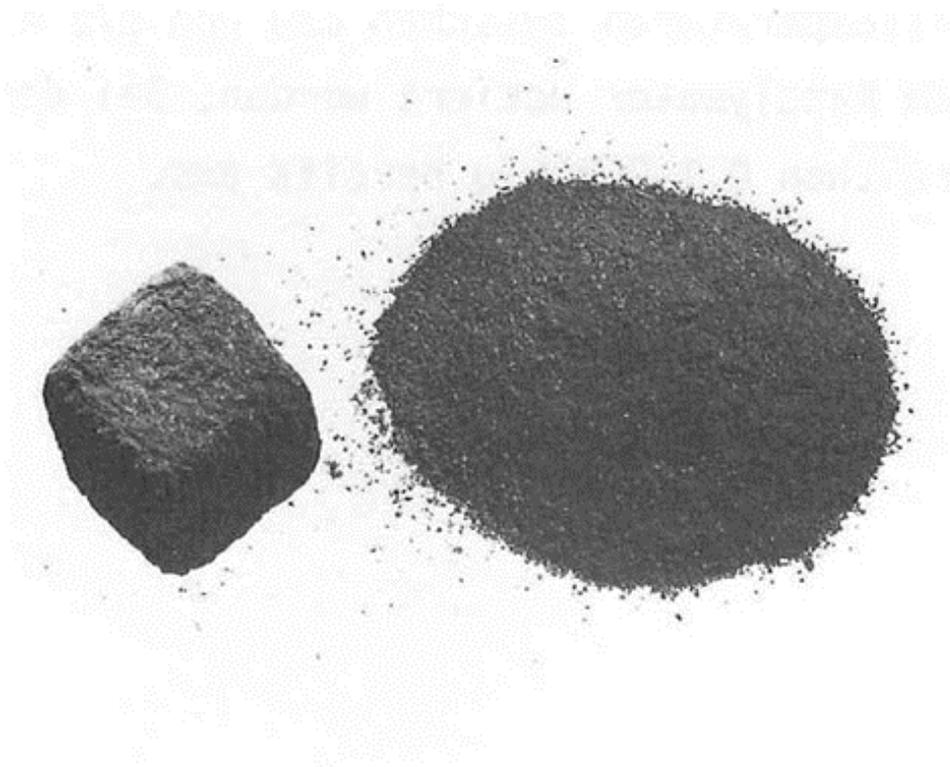


Figure 45: Corroded A3-matrix material (from Hinssen *et al.* 1982)

### 10.12 A3-Matrix Disintegration by Bromine Vapor

The Seibersdorf Research Center in Austria pioneered the development of processes to retrieve the irradiated BISO and/or TRISO fuel particles from the A3-matrix, without damaging the particle layers and proving their fission-product retention capabilities (Reitsamer *et al.* 1985).

A variety of tests were conducted to disintegrate the A3-matrix material under remote hot cell conditions. This includes reactions of A3-matrix material with bromine, bromine-iodine, iodine-chlorine, alkali metals, and metal halogenides such as aluminum chloride, ferric chloride etc.

Bromine vapor was used as a candidate to intercalate into the graphite lattice and widen the *c*-axis in a way that the crystals lose their mechanical strength and thus enabling fragmentation.

Figure 45 shows a fuel pebble treated with bromine vapor for about one hour at 350-400°C. No attack of the coated particle layers was observed as the pyrocarbon shows no intercalation effects. Bromine was retrieved by a freezing process. The small portion of bromine bound to the graphite powder by adsorption was driven off by heat treatment at 300°C.

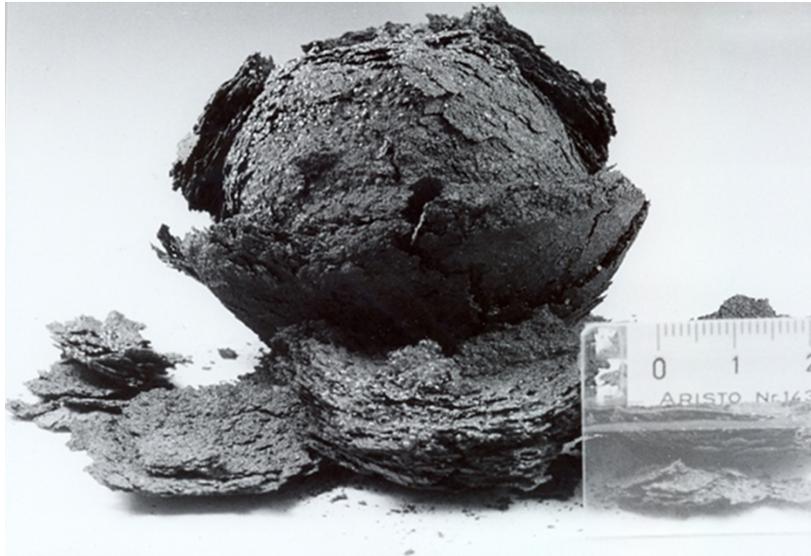


Figure 45: Fuel pebble disintegrated by bromine vapor (HTR Archive, FZJ Jülich, Germany)

*‘The advantage of this method is the simplicity of the procedure and a completely dry end product. One of the disadvantages is that the resulting graphite powder is coarse grained; as a result of this coarse texture, the coated particles are sometimes not completely freed of matrix graphite, with the residual graphite continuing to adhere to the particle coating as a so-called "over-coating". The removal of this over-coating requires a further process step, e.g., boiling with carbon tetrachloride’ (Bildstein and Knotik 1973).*

Further difficulties arise from the aggressive reactive nature of bromine with regards to the protection of sensible instrumentation and hot cell manipulators.

### 10.13 Intercalation by Acids

Intercalation of acids<sup>22</sup> into the crystal lattice has been adapted as another technique for the disintegration of fuel particles. Guittonneau (2010) used low ( $\text{H}_2\text{SO}_4 + \text{H}_2\text{O}_2$ ) and high ( $\text{H}_2\text{SO}_4 + \text{HNO}_3$ ) temperature acid treatments. The fuel compacts contained about 3000 TRISO particles with  $\text{ZrO}_2$  (dummy) kernels and a packing fraction of 20%.

A variety of Graphite Intercalation Compounds (GICs) can be generated by using intercalates such as alkali, earth alkali, transition metal chlorides, acids, halogens, etc., in gas-solid or liquid-solid phase (Guittonneau 2010). GICs are often used as an interim product before starting an exfoliation process with a large expansion of the graphite crystals (Exfoliated Graphite (EG)) and high surface ratios ( $\sim 85\text{m}^2/\text{g}$ ) (Figure 46).

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<sup>22</sup> Illustrations, results and interpretations in this section are provided by courtesy of SUBATECH, France

Gas formation by the disintegration of hydrogen peroxide and the related release of oxygen contributed to the volume increase and supported the extraction of the TRISO particles. Thus, there is no need for the formation of high stage GICs if the separation of TRISO particles is the primary aim. TRISO particles and the GICs/EGs have been separated by a flotation process.

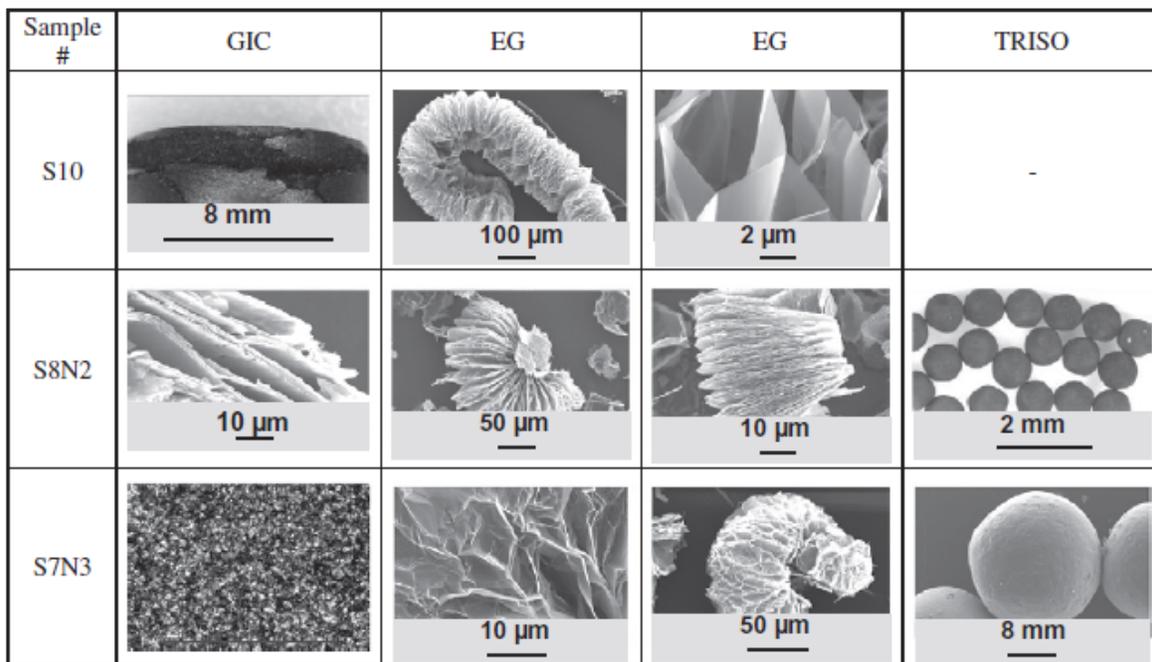


Figure 46: GIC, EG and the extracted TRISO particles (from Guittonneau et al. 2010, courtesy SUBATECH France)

The GICs have been heated within one min to 1000°C for decomposing acids and generating the EGs, which are quite oxidation resistant.

Room temperature treatments with different ratios of  $H_2SO_4$  and  $H_2O_2$  started with a rather quick swelling within several minutes, as can be seen in the following Figure 47:

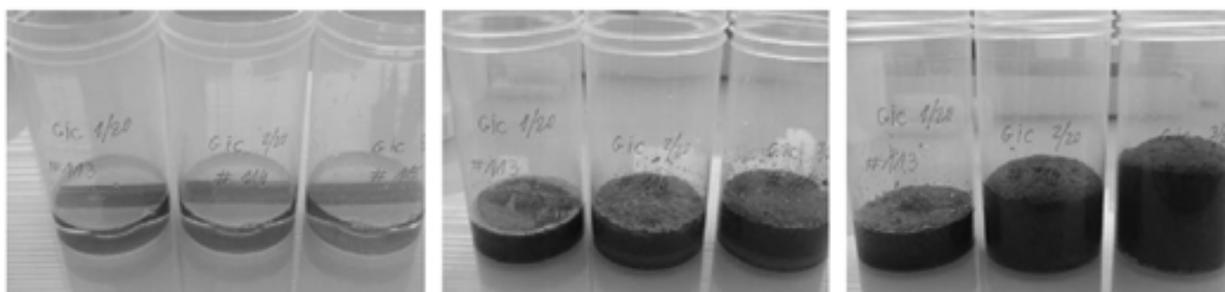


Figure 47: Expansion at time  $t = 0$ ,  $t = 9$  and  $t = 17$  min after mixture of  $H_2SO_4$  into  $H_2O_2$  (from Guittonneau et al. 2010, courtesy SUBATECH France)

GICs and EGs could however be interesting for successive purification from radioisotopes and further industrial reuse of the residual carbon material. Compressed GICs are, for example, used to manufacture flexible graphite foils (Enoki et al. 2003).

## 10.14 Intercalation by Electrochemical Methods

A high degree of intercalation by electrochemical methods is easier to achieve than by oxidizing acids alone: ‘When graphite is electrochemically oxidized, anions are intercalated to compensate for the positive charge generated in graphite. One of the advantages of the electrochemical method is the possibility to control the structure and composition of the resulting intercalation compounds by changing the applied potential or the degree of charging’ (Matsuo *et al.* 2024).

Over the past decades of HTGR fuel development, electrochemical processes (e.g. Figure 48) have been applied in the related PIE tests (Verfondern and Nabelek 2017).

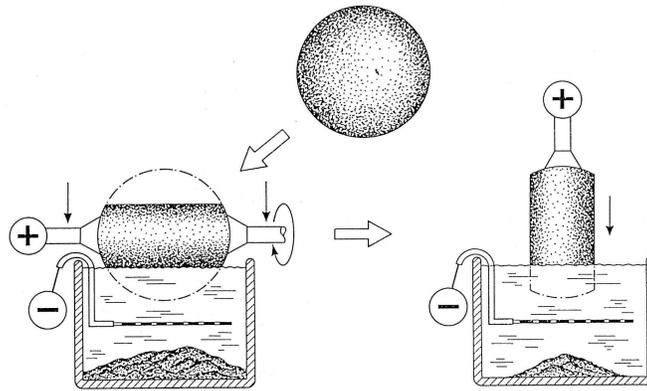


Figure 48: Electrochemical decomposition of HTGR fuel pebbles (from Schenk *et al.* 1986)

Investigations on the electrochemical decomposition of HTGR fuel pebbles showed that an increased amount of GC binder in the A3-matrix material augmented the chemical oxidation, with more pronounced production of graphite oxide (Zhang *et al.* 2019).

Improved electrochemical methods with molten salts as electrolytes keep the advantages of other electrochemical methods and avoid the problems caused by severe oxidizing acids (Tian *et al.* 2009).

Investigations on the electric current intensity show an influence on the released carbon mass and carbon particle size. Higher electric currents tend to damage the outer PyC layers of extracted TRISO particles (Chen *et al.* 2017).

In summary, intercalation and electrochemical methods have already been applied on PIE on irradiated HTGR fuel pebbles and compacts for investigating the integrity and fission product retention capabilities of BISO and TRISO particles, in hot-cell laboratories. The processes will still need to be extrapolated to a higher throughput for waste volume reduction purposes. The corrosive nature of the applied chemicals exhibits a challenge for the exposed equipment. Molten salt processes may alleviate the handling of the processes, which still needs to be demonstrated on a semi-technical scale. Secondary waste is also corrosive and needs to be neutralized before being purified and disposed.

Homogeneous oxidation is an alternative option to weaken the A3-matrix without endangering the integrity of the TRISO particles. Thus, homogeneous oxidation may also be used as a pre-treatment with minimal secondary waste streams.

## 11 Summary

The management of irradiated graphite waste from nuclear reactors presents a complex and multifaceted challenge, and future designs may or may not use a comprehensive and integrated approach. This report has highlighted several key considerations and strategies for effective and safe management of this type of waste.

### Dose and Volume Reduction

One of the primary goals in managing irradiated graphite waste is to reduce both the radiation dose and the volume of waste. Various strategies have been investigated, including:

- **Delayed Dismantling:** Allowing short-lived isotopes to decay before dismantling can significantly reduce the radiation dose. However, this approach must be balanced against the risk of losing institutional knowledge and expertise over time.
- **Volume Reduction Techniques:** Techniques such as mechanical fragmentation, thermal treatment, and chemical processing can reduce the volume of waste, making it easier and more cost-effective to handle and dispose of.

### Recycling and Reuse Opportunities

Recycling and reuse of irradiated graphite can provide significant benefits, including the recovery of valuable isotopes for medical and industrial applications. Potential recycling strategies include:

- **Thermal Treatment:** Heating graphite to release isotopes such as tritium and  $^{14}\text{C}$ , which can then potentially also be captured and reused.
- **Chemical Processing:** Using chemical methods to extract valuable isotopes and reduce the overall radioactivity of the waste.

### Design Considerations

The designs of new reactors may involve incorporating features that facilitate the eventual dismantling and disposal of graphite components. This includes the possibility that licensees may select graphite materials that are less prone to activation and designing reactor components to minimize the generation of radioactive dust and debris.

### Lessons from High-Temperature Gas-Cooled Reactors (HTGRs)

Much of the information in this report is based on experience with HTGRs. Key lessons from these reactors include:

- **Graphite Behavior:** Understanding the physical, chemical, and mechanical behavior of graphite under irradiation enables planning its management.
- **Dust and Debris Management:** Effective strategies for managing radioactive dust and debris may be utilized to minimize contamination and facilitate safe handling and disposal.

## Molten Salt Reactors (MSRs)

While there is relatively little information available on graphite waste management for MSRs, ongoing research is expected to provide valuable insights. Key areas of investigation include:

- **Graphite-Salt Interactions:** Understanding how molten salts interact with graphite and how this affects the behavior and management of graphite waste. Research organizations are presently investigating these interactions.
- **Waste Treatment and Disposal:** Developing effective methods for treating and disposing of graphite waste from MSRs. There is little existing experience in this area.

This report helps prepare the NRC staff for efficient licensing reviews of graphite waste storage and transportation for the increased and differing graphite wastes expected from advanced reactors.

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