CP-68001
Revision 0

Review of Potential Wigner Effect Impacts on the Irradiated Graphite in Decommissioned Hanford Reactors

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Contractor for the U.S. Department of Energy
under Contract 89303320DEM000030

CPCCo
Central Plateau
Cleanup Company

P.O. Box 1464
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Terms

BGRR	Brookhaven Graphite Research Reactor
FEP	features, events and processes
R&D	research and development
UK	United Kingdom

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1 Introduction

This review of is motivated by the need to consider future disposal of the nine surplus Hanford Site production reactors (B, C, D, DR, F, H, KE, KW, and N) that are decommissioned as part of the Hanford Site Composite Analysis. The B Reactor has been designated as a museum. The other eight reactors will be evaluated in a future performance assessment before a final disposal facility can be authorized to construction or to receive this waste form. After a performance assessment is available, then the Hanford Site Composite Analysis would be updated to account for this additional source term. Disposal is currently assumed to occur in calendar year 2070 by one-piece removal to a projected disposal facility in the 200 West Area of the Hanford Site Central Plateau. The N Reactor core may also be disposed analogously to the single-pass reactors. It will further assume that the B Reactor will remain a museum permanently. The U.S. Department of Energy studied at least five decommissioning alternatives for long-term, safe management of radionuclide-contaminated materials inside the eight single-pass reactors (DOE/EIS-0119-FEIS, *Decommissioning of Eight Surplus Production Reactors at the Hanford Site, Richland, Washington*). Note that N Reactor, a closed-loop reactor, was not addressed in DOE/EIS-0119-FEIS. The five decommissioning alternatives evaluated were as follows:

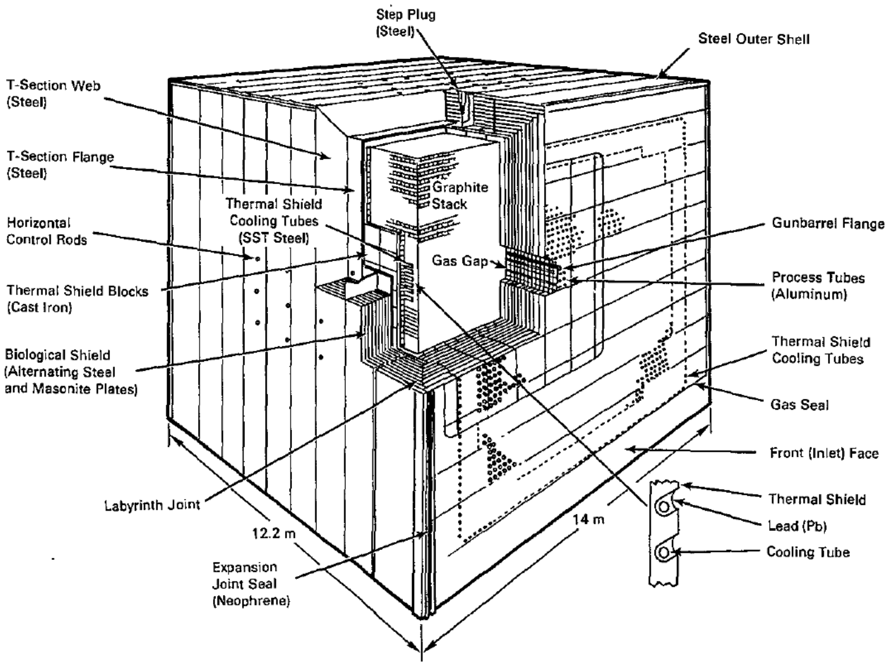
1. No action alternative
2. Immediate one-piece removal alternative
3. Safe storage followed by deferred one-piece removal alternative
4. Safe storage followed by deferred dismantlement alternative
5. In situ decommissioning alternative

DOE/EIS-0119-FEIS considered environmental consequences, socio-economical resources and land uses, costs, and statuary and regulatory requirements. Once an alternative is determined, detailed performance assessments will be conducted to further confirm the safety of the long-term disposal of the eight reactors.

To achieve the goal of long-term safety of the chosen decommissioning alternative, any factors that have the potential to affect the safety must be assessed. This report examines the potential impact from one of such factors (i.e., the residual energy stored in the irradiated reactor graphite).

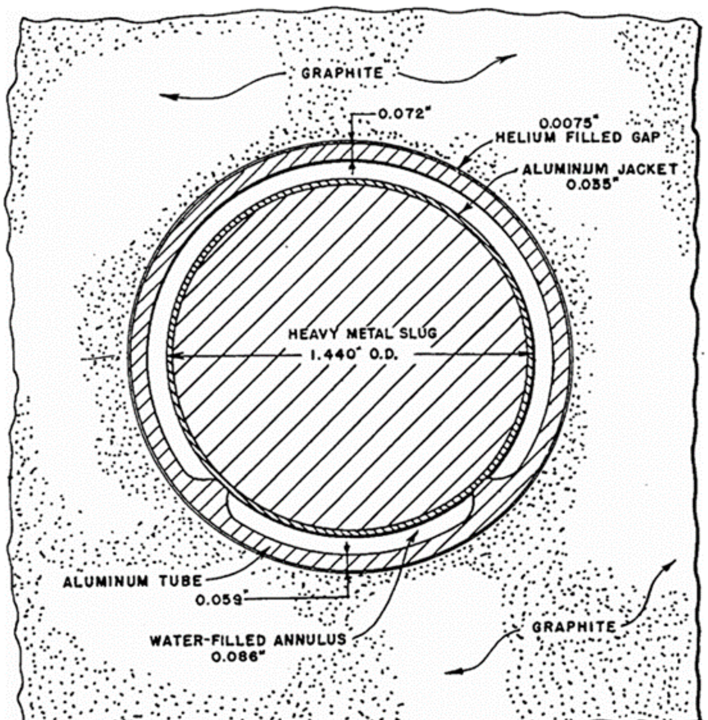
The surplus production reactors at the Hanford Site used graphite as the moderator to reduce fast neutrons to thermal neutrons that can trigger fission reactions in fissile radionuclides such as uranium-235. The graphite block combined with the thermal shielding in a typical Hanford Site reactor to be decommissioned is about $14\times12\times14$ m^3 and 11,000 tons (DOE/EIS-0119D, *Decommissioning of Eight Surplus Production Reactors at the Hanford Site, Richland, Washington*) as shown in Figure 1. Each individual graphite block is about 0.11×0.11 m^2 and 1.21 m long. Each graphite block is penetrated by an aluminum tube containing metallic uranium fuel slugs that is enriched with 0.96 without uranium-235 as shown in Figure 2. These figures illustrate that as the graphite performed to slow down fast neutrons, the material was bombarded by the energetic particles during reactor operation.

The irradiation by fast neutrons, in particular, would displace atoms from their normal positions to cause lattice defects in the carbon network and produce interstitials between the crystal planes of the graphite. The consequence is an increase of the internal energy of the graphite. The increased internal energy, or the stored energy, termed “Wigner energy,” is inherently unstable, and the affected graphite has the tendency to minimize, or release, the energy upon encountering the triggering conditions, such as heating. If the energy is released suddenly, it could cause rapid rise in temperatures locally.



Source: DOE/EIS-0119D, *Decommissioning of Eight Surplus Production Reactors at the Hanford Site, Richland, Washington*.

Figure 1. Illustration of the Graphite Block in a Typical Legacy Graphite-Moderated Legacy Reactor



Source: Reed, 2019, "The Hanford Engineer Works."

Figure 2. Cross-Sectional Illustration of the Graphite-Uranium Fuel in a Typical Legacy Graphite-Moderated Reactor

For the decommissioned Hanford Site surplus production reactors, whatever the decommissioning alternative to be decided, the sudden release of the energy stored in the graphite can also raise concerns. This report covers the literature review of the existing publications and presents the knowledge status, from which the future work or actions, if any, are recommended. The literature reviewed are grouped into the following categories:

- Impact on waste disposal safety – This category (e.g., NuSAC, 2005, *Meeting of RG2 with Windscale Pile 1 Decommissioning Project Team 29/09/2005*; Research and Development (R&D) Technical Report P3-80/TR, *Wigner Energy in Irradiated Graphite and Post-Closure Safety*; EPRI, 2006, *Graphite Decommissioning, Options for Graphite Treatment, Recycling, or Disposal, including a discussion of Safety-Related Issues*; Guppy et al., 1999, "Technical Assessment of the Significance of Wigner Energy for Disposal of Graphite Wastes from the Windscale Piles"; Minshal and Wickham, 1999, "The Description of Wigner Energy and its Release from Windscale Pile Graphite for Application to Waste Packaging and Disposal"; IAEA-TECDOC-1154, *Irradiation Damage in Graphite due to Fast Neutrons in Fission and Fusion Systems*; IAEA-TECDOC-1647, *Progress in Radioactive Graphite Waste Management*; IAEA-TECDOC-1790, *Processing of Irradiated Graphite to Meet Acceptance Criteria for Waste Disposal*) contains review and reports the past and up-to-date R&D of Wigner energy stored in the irradiated graphite, concerns arising from the previous Windscale accident that is suspected to be caused by the sudden release of Wigner energy, potential consequences on the decommissioning and disposal of reactor graphite waste.
- Impact on reactor safety – This category (e.g., NUREG/CR-4981, *A Safety Assessment of the Use of Graphite in Nuclear Reactors Licensed by the U.S. NRC*; ORNL/NRC/LTR-09/03, *Milestone Report on the 'Workshop on Nuclear Graphite Research'*; INL/EXT-07-13165, 2007, *Graphite Technology Development Plan*; IAEA-TECDOC-901, *Graphite Moderator Lifecycle Behavior*; Nordlund et al., 2018, "Primary Radiation Damage: A Review of Current Understanding and Models"; Pavliuk et al., 2019, "Dynamics of Temperature fields During Wigner Energy Release in Bulk Graphite Irradiated at Low Temperature"; and BNL-77205-2006-IR, *Analysis of Wigner Energy in BGRR Graphite, Final Analysis*) studies the impact on reactor safety arising from radiation damage to graphite. The literature contains many data that help reveal the stored energy in Hanford Site reactor graphite moderators.
- Modern material research – This category (Ma, 2007, "Simulation of Interstitial Diffusion in Graphite"; Li, 2005, "Defect Energies of Graphite: Density-Functional Calculations"; Gulans et al., 2011, "Bound and free self-interstitial defects in graphite and bilayer graphene: A computational study") provides a better and new understanding on the energy stored and the dynamics of the interface between the graphite lattice and interstitials, which provides an insight on the "fate" and the spectrum of the stored energy.

The review findings are presented in the following sections. First, the investigation result of the Windscale accident will be briefly reviewed. Secondly, the new understanding of the Wigner effect will be presented. This will help improve understanding of the experimental data on annealing and energy release. Thereafter, a summary of parameters and conditions associated with the release of the stored energy will be presented. This is followed by the review of potential effects on the decommissioning and disposal of the radioactive graphite waste. At the end of the report, conclusions and recommendations will be presented.

2 Windscale Accident Investigation

This chapter summarizes the main findings of the investigation of the Windscale Pile No.1 accident (R&D Technical Report P3-80/TR). This accident happened in 1957 and involved a fire during a controlled heating process intended to release the stored energy in the graphite within an operating reactor (Windscale Pile No.1). After the incident, the sudden release of Wigner energy in the graphite moderator was suspected as the culprit of the fire. After this and the later Chernobyl accident, there was an increased interest in the fire risk arising from the irradiated graphite burning among scientific and engineering communities of nuclear waste management and reactor design and operation. In the field of nuclear waste management, the sudden release of the stored energy from the irradiated graphite is considered as features, events, and processes (FEPs) in the safety assessment (R&D Technical Report P3-80/TR).

After investigation of the accident, the United Kingdom Nuclear Safety Advisory Committee concluded in 2005 (NuSAC, 2005) that:

> ……there was NOT a graphite fire: damage to graphite, caused by severely overheated fuel assemblies, was localized…

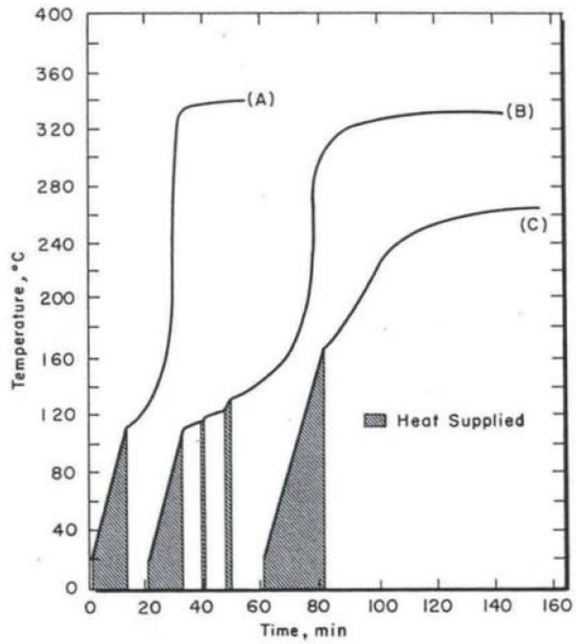
More detailed and technical descriptions of both the accident and conclusion to exclude the connection between the fire and the release of stored energy can be found in NUREG/CR-4981. The study concluded the following:

> It is shown that there is no evidence from the Chernobyl event that stored energy releases played a role either initiating or contributing to this accident. A controlled process of annealing the graphite at Windscale with nuclear heat resulted in damage to the fuel elements that initiated fuel burning which resulted in a graphite fire. Stored energy releases did not initiate or contribute to this accident either.

This report (NUREG/CR-4981) further stated that because the graphite burning temperature is 650°C while the uranium fuel in Windscale Pile No.1 is metallic with a burning temperature of 300°C (under oxygen-rich environment and without, or with, failed cladding), it is pertinent that the uranium fuel would burn before the graphite could burn. The graphite must be heated to and maintained at 650°C to initiate burning and to sustain a fire, which excludes the burning of the graphite as a credible scenario in the storage and disposal of the surplus production reactors. More detailed description on the graphite burning tests can be found in EPRI (2006), which also supports the above-mentioned U.S. Nuclear Regulatory Commission conclusion. At the latest international nuclear graphite specialists' meeting, the experts expressed the need to convince regulators that "the graphite does not burn."[1]

Nevertheless, the release of the stored energy remained a concern in safe management of the decommissioned graphite-moderated reactors. For example, temperatures of the irradiated graphite were shown to rise rapidly after initial heating period under adiabatic (with no effective heat transfer) conditions shown as "adiabatic rise" curves in Figure 3. The figure shows that once the graphite is heated to a certain temperature, the energy released from the graphite would provide heat to sustain further energy release under adiabatic conditions. For example, the initiation temperature would be less than 120°C for sample A and would be as high as 160°C for sample C. This temperature is called initiation temperature of self-induced Wigner energy release by Pavliuk et al. (2019).

[1] Unlike coal that contains organic materials and is porous, graphite is tightly packed atomic-scale sheets and has very little specific surface area for oxidation.



Source: ORNL/TM-2011/378, *A Review of Stored Energy Release of Irradiated Graphite*. Curve B illustrates the way in which the starting rate can be changed until a suitable rate is obtained.

Figure 3. Adiabatic Rise Curves for Typical High Energy Sample A and Typical Low-Energy Sample C

3 Review of Wigner Effect Research

Research on Wigner effect started in 1940s soon after the effect was discovered. Since then, two stages in research can be identified. This section briefly summarizes the early stage (1943 through 1970s) research and reviews the relevant research results in the current stage (since 2000s).

3.1 Early Studies

The Wigner effect includes the stored energy and altered graphite properties. When first discovered in 1942, the strains of the interstitials accumulated between the lattice layers (Simmons, 1965, *Radiation Damage in Graphite*) were identified as the main issues in reactor safety and performance:

> …fast neutrons would displace atoms from their normal positions and so produce lattice defects in the form of holes in carbon networks and interstitial atoms intercalated between the layer planes of the graphite. The interstitial atoms would cause an increase in the interlayer spacing and so cause the graphite to grow. Lattice strains produced by the defects would increase the internal energy of the graphite.

While vacancies tend to reduce the stored energy by pairing with the displaced atoms (the annealing process), interstitials are the primary concern. Before the Windscale Pile No.1 accident in 1957, study of Wigner energy was focused on the radiation damage to the graphite moderator such as changes in dimension, thermal, mechanical, and electrical properties. The general understanding is that the release of the stored energy requires the interstitials to become mobile, which requires more energy to overcome the mobility barrier. If the energy supplied to the irradiated graphite is sufficiently high, the stored energy can be released. For graphite inside an operating reactor, heating is considered as an efficient way to supply the annealing energy and had been successfully carried out to a large scale (IAEA-TECDOC-1790). Heating that caused the accident in Windscale Pile No.1 was intended to induce a controlled annealing.

The emphasis of the study was on thermal annealing of the stored energy of the graphite irradiated at low temperatures (less than approximately 150° to 250°C). The energy release is associated with temperature rise of the studied graphite specimens (ORNL/TM-2011/378, *A Review of Stored Energy Release of Irradiated Graphite*). These studies (reviewed and published in Guppy et al., 1999; Minshall and Wickham, 1999; and ORNL/TM-2011/378) generated many experimental data and provided the following observations for the graphite irradiated at low temperatures:

- The temperature required to release the energy is proportional to the temperatures of irradiation and the irradiation dose.
- There is a range of activation energy, the energy required to trigger the release of the stored energy.
- The total stored energy increases with irradiation dose and appears to reach a saturation limit after long exposures.
- The rate of accumulation of stored energy and the saturation value decreases with increasing irradiation temperature.

At least one of these experimental studies used the Hanford Site reactor graphite as shown in Figure 4 that shows the stored energy release rate as a function of annealing temperature for different specimens irradiated at various neutron fluences[2]. These curves show that the energy release rates for the specimens irradiated at the neutron fluence less than 10×10^{-19} neutron per cm^2 (n/cm^2) peak at about 200°C. These specimens appeared to have released most of the stored energy when the annealing temperature reached 500°C. The specimens irradiated with the neutron fluence greater than 10×10^{-19} n/cm^2 may need higher annealing temperatures to release most, if not all, of the stored energy. More experiments observed similar peaks at 200°C for releasing the stored energy in the graphite irradiated below 150°C.

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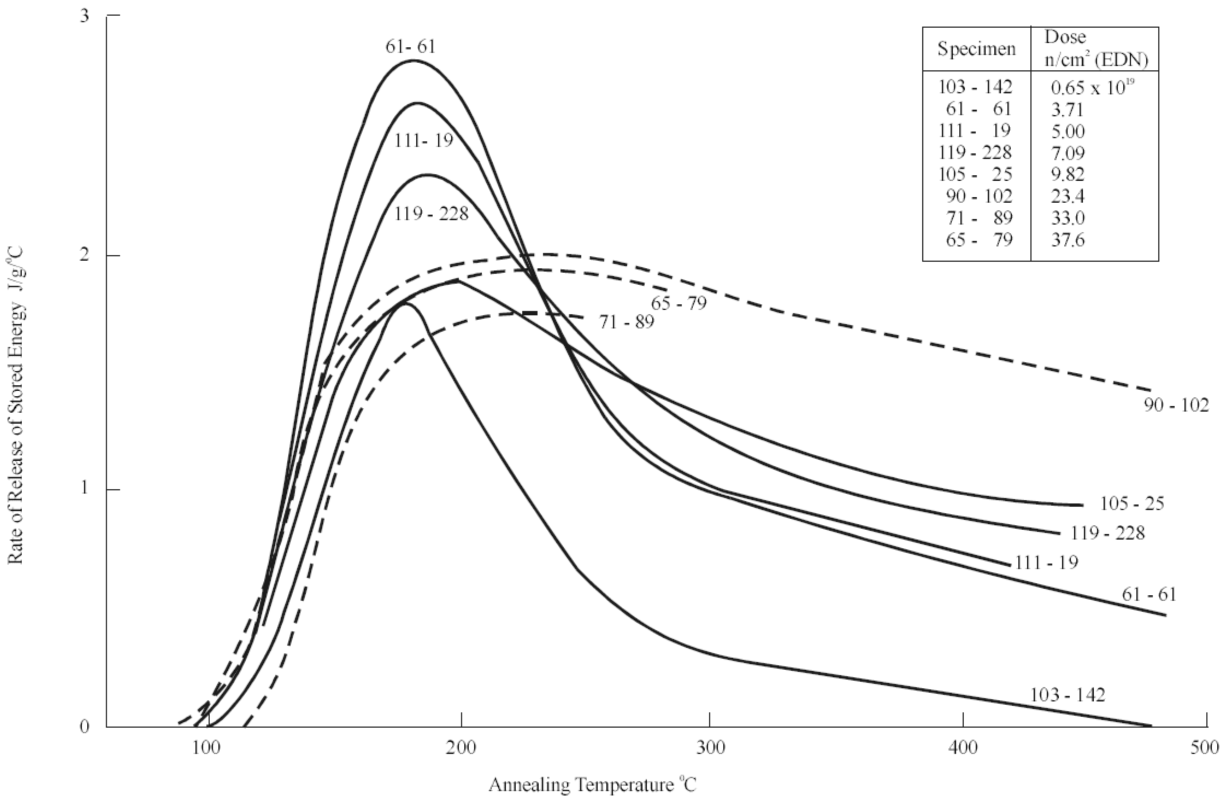
[2] Fluence in the context of the report has a unit of neutrons/area (n/cm^2 or n/m^2). In some literatures (e.g., IAEA-TECDOC-1521, *Characterization, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Reactors*), "fluence" is used to describe neutron flux. In other literatures (e.g., INL/EXT-07-13165, *Graphite Technology Development Plan*), "fluence" is equivalent to dose measuring displacement damage (i.e., dpa – displacements per atom in solids). A conversion given by INL/EXT-07-13165 is 0.78×1021 n/cm^2 = 1 dpa. In some literatures (e.g., IAEA-TECDOC-901, *Graphite Moderator Lifecycle Behavior*) the same unit of n/cm2 is used for "neutron dose."



Source: IAEA-TECDOC-1154, *Irradiation Damage in Graphite due to Fast Neutrons in Fission and Fusion Systems*.

Figure 4. Stored Energy Release Curves for Graphite Irradiated at Approximately 30°C in the Hanford Site K Reactor Cooled Test Hole

The early modeling studies used Boltzmann distribution to derive the activation energy or activation temperature. An example of the expression in terms of release rate of stored energy (R&D Technical Report P3-80/TR) is:

$$\frac{dS}{dt} = \upsilon S(E,t)\exp\left(-\frac{E}{kT}\right) \quad \textbf{(Eq. 1)}$$

where:

S = stored energy (expressed in eV or equivalent energy unit)

t = time

υ = frequency factor

E = activation energy (eV)

k = 8.617×10^{-5} (eV/K) is the Boltzmann constant

T = absolute temperature (K).

The frequency factor υ is in general on the order of magnitude of the vibrational frequency of an atom on a lattice. In systems concerned in this report, this parameter represents vibrational frequency of a vacancy or an interstitial attempting to move away from its current position. The exponential term represents the probability of the vacancy/interstitial moving away from its current position by overcoming the energy barrier. The average energy barrier can be represented by E, the activation energy, in Equation 1. This means that if the activation energy is high, the annealing temperature must also be sufficiently high to release the sufficient amount of the stored energy.

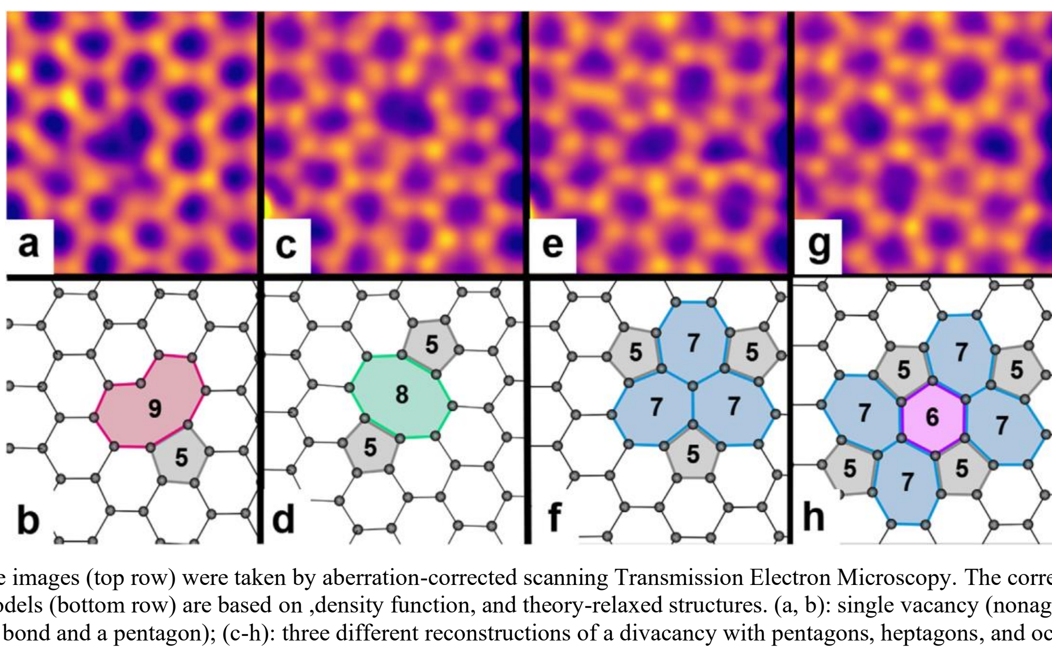
Parameters in the Boltzmann distribution expressions can be determined from measured data. These parameters were helpful in interpreting the experimental data and were expected to provide assistance to thermal annealing design. Nevertheless, the Boltzmann distribution does not reveal the microscopic nature of radiation-induced defects on theoretical level.

3.2 Modern Studies

Fundamental R&D related to Wigner effect emerged in the late 1990s in material sciences because of the growing interest in graphitic-based material applied in nano-technologies. These materials are of major technological interest, and radiation effects have to be taken into account in research and applications in space and zones of high radiation level such as nuclear materials in advanced reactors, as well as utilizing radiation to tailor the desired nanomaterials. Advanced microscopic experimental studies and theoretical modeling help improve understanding of Wigner effect. On the theoretical level, in particular, first-principles (quantum mechanics) modeling enables studying intrinsic defect phenomena in real atomic systems.

Among these materials, graphene (single atomic layer of graphite) stands out as an ideal system to study atom displacements in detail, to test the theoretical concepts, and to determine threshold energies with high precision (Nordlund et al., 2018). Because the displacement of even single carbon atoms can be observed in situ in the electron microscopes, the situation in graphene is now very well investigated and understood. On the other hand, electron irradiation leaves artifacts in electron microscopes, which must be avoided. This requires detailed knowledge of irradiation defect formation and annealing.

Figure 5 shows an example of measurement and theoretical simulation of divacancies (the space left by two displaced atoms) in irradiated graphene as an example to demonstrate the affinity between experimental (top) and theoretical (bottom) studies.



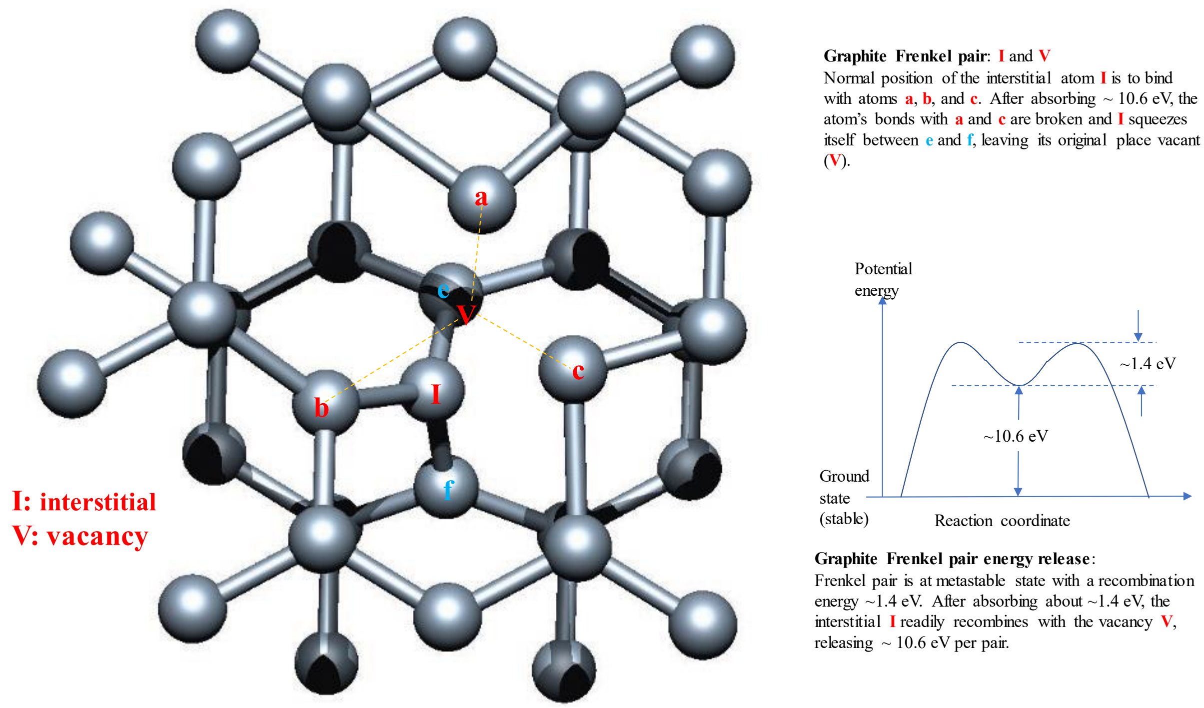
Note: The images (top row) were taken by aberration-corrected scanning Transmission Electron Microscopy. The corresponding lattice models (bottom row) are based on ,density function, and theory-relaxed structures. (a, b): single vacancy (nonagon with one open bond and a pentagon); (c-h): three different reconstructions of a divacancy with pentagons, heptagons, and octagons (Nordlund et al., 2018, “Primary Radiation Damage: A Review of Current Understanding and Models”).

Figure 5. Different Configurations of Vacancies in a Monolayer of Graphene

The modern material R&D reveals that although Wigner's theory of stored energy arising from interstitial caused strains could explain observation to a certain degree, more microscopic phenomena and defect dynamics, in particular, the atomic bond, are responsible for storing and releasing of irradiation energy. Relevant graphite defect studies are briefly reviewed below.

3.2.1 Frenkel Pair

Using first-principles theory, Telling et al., 2003, "Wigner Defects Bridge the Graphite Gap," modeled the Frenkel defects in the irradiated graphite moderator. The Frenkel pair (Figure 6) is believed to be the primary source of Wigner energy (Ewels et al., 2003, "Metastable Frenkel Pair Defect in Graphite: Source of Wigner Energy?"). Telling et al. (2003), predicted an energy barrier for recombination of a Frenkel pair to be 1.4 eV that is in agreement with experimental observations on the peak release rates of the stored energy occurring at about 200°C for graphite irradiated at low temperatures (such as Figure 7). The strain energy caused by the interstitial in Frenkel pair is only a fraction of the recombination energy of approximately 1.4 eV.



Source for left graphic Telling et al., 2003, "Wigner Defects Bridge the Graphite Gap."

Figure 6. A Frenkel Pair in Graphite (left) and Illustration of its Stored Energy (Approximately 1.6 eV) and the Barrier (Approximately 1.4 eV) for Releasing the Stored Energy (right)

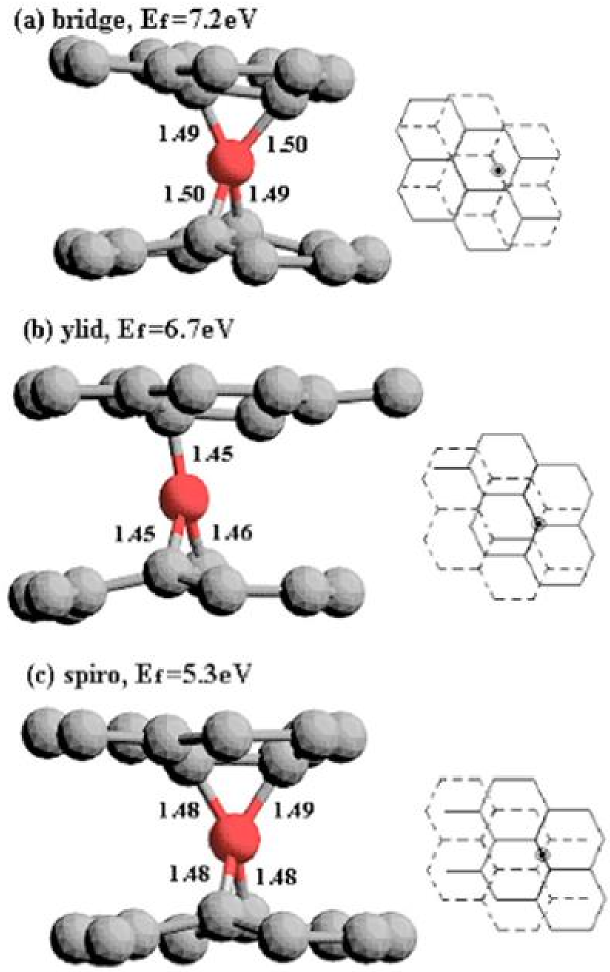


Figure 7. Examples of Three Interstitials (red ball) Between Two Sheared Graphite Planes (consisting of gray atoms) Along with Formation Energies E_f and Bond Lengths in Angstrom (10^{-10} m)

3.2.2 Migration of Vacancies

Vacancy defects take several configurations. Divacancies shown in Figure 5 are one of these configurations and believed to be the important annealing mechanism in graphitic materials (Nordlund et al., 2018). The divacancies are formed in two ways. First, monovacancies (formed by one atom displacement) have very low migration energies (0.1 – 1.4 eV) and can be mobile at temperatures slightly above room temperatures (determined from expressions similar to Equation 1). The monovacancies then coalesce to form divacancies. Secondly, divacancies can form directly from irradiation displacement because they have similar formation energies (energies to displace two atoms 7.2 – 7.9 eV) as monovacancies (7.3 – 7.5 eV). Compared to monovacancies, divacancies are stable and immobile (with migration energy approximately 7 eV) unless at very high temperatures. This migration energy might correspond to the peak of release rate of stored energy at high temperatures (>1200°C) reported in several literatures (e.g., ORNL/NRC/LTR-09/03).

Besides divacancies, monovacancies may diffuse to join small vacancy clusters. Some of the vacancy clusters have much lower migration energies (0.95 to 1.93 eV) compared to divacancy (El-Barbary, 2018, "Vacancy Cluster in Graphite: Migration Energy and Aggregation Mechanism").

3.2.3 Migration of Interstitials

Besides Frenkel pair and vacancies, diffusion of interstitials also contributes to the release of the stored energy. Most relevant studies (Li et al., 2005; Ma, 2007; and Gulans et al., 2011) used first-principles to calculate the migration barriers of diffusion of interstitials in various states. Primarily, these interstitials can be classified into "free" and "bound" states. "Free" interstitials are single atoms that do not bound with other atoms including lattice atoms and interstitials. "Free" interstitials have very low migration barriers, typically in the range of 0.1 – 0.5 eV, which means these interstitials may be mobile at room temperatures or even lower. These results are consistent with the low-temperature self-diffusion of interstitials in graphite that has been experimentally observed for decades. Note that these self-diffusing interstitials may not all resolve to stable states, i.e., recombine with vacancies. They may form clusters and become "bound" interstitials, depending on formation energies of the various "bound" configurations and their formation energies, locations of the interstitials, and pre-existing defects (next section).

"Bound" interstitials take various configurations. Interstitials in Frenkel pair (Figure 6) can be considered as a special form of "bound" interstitial configuration. "Bound" interstitials between lattice planes may take various configurations and their formation energies ranges from 6 – 10 eV according to calculations using different first-principles models. If there is a shear force between the two graphite planes, the interstitials between the planes tend to bound more lattice atoms on the planes, which is more stable (or less stored energy or less mobile) than those without shear force. The inter-plane interstitials also tend to migrate in the form of exchanging lattice atoms. For bound interstitials, the migration energies are found to be greater than 1.2 eV. Figure 7 shows three examples of configurations of interstitials between two graphite planes with shear forces.

3.2.4 Preexisting Defects

Studies also find that defects existed prior to irradiation affect the stored energy arising from radiation induced defects (Nordlund et al., 2018). Real materials contain dislocations, grain boundaries, and various other defects. Surfaces also influence the rate of radiation defect production. Most significantly, even if the initial crystal structure of a materials were ideal, the generation of damage itself from the previous irradiation results in the gradual defect accumulation and dislocations. Grain boundaries tend to be sinks for interstitials to settle in where some of their energies are reduced because the grain boundary has a sufficient space to cause no or little strains to the bulk graphite. Dislocations tend to cause vacancy clusters or loops (one-by-one linked).

3.2.5 Macroscale and Microscale Studies

In this section, the "macroscale" study refers to nuclear scientists' efforts to understand Wigner energy and release Wigner energy through annealing. Macroscale studies energies stored in bulk graphite after irradiation. The "microscale" study in this section refers to material scientists' efforts to explore individual or a cluster of defects caused by irradiation. Microscale focuses on types of defects and their configurations including interactions with bulk graphite through graphite lattice planes. There is apparently a linkage, for example, defect migration energies in microscale and activation energies in macroscale. Nonetheless, the two fronts proceed separately due to lack of common goals.

4 Parameters Relevant to Stored Energies

Modern material research provided new insights of the energy storage in graphite associated with neutron irradiation in reactors. These models and experimental methods, however, cannot quantitatively study the irradiated graphite in engineering scales, and hence cannot provide engineering parameters for managing the stored energies. Nevertheless, qualitative information can be extracted from these studies to aid management of the stored energies.

4.1 Formation Energies

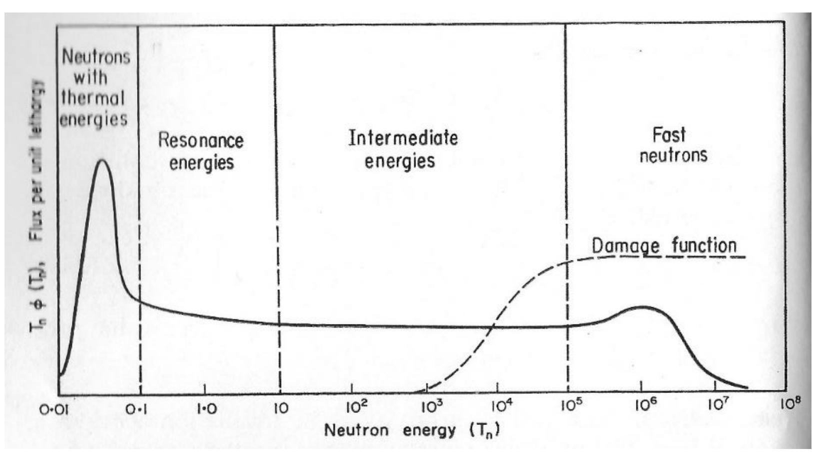
Formation energies of various configurations of interstitials provide two pieces of information. First, knowing the formation energy of a defect configuration or structure helps determine the possibility of formation of such defect configuration or structure. Secondly, the formation energy of a defect determines the amount of stored energy, and hence the release of the stored energy for a given type of defect. Depending on the types or configurations of a defect, the formation energy may range from 5 to 10 eV.

4.2 Migration Energies

Migration energies refer to the diffusion barriers of interstitial and vacancy or the clusters of them. This parameter is important to annealing because the interstitials and vacancies must have kinetic energies greater than the migration energies to initiate diffusion. Once in motion, these will recombine to settle in stable states and release the stored energy. Most migration energies for the defects are around 0.1 to 1.5 eV, corresponding to annealing temperatures of around room temperatures to several hundreds of Celsius degrees. Some defects, such as divacancies have a migration energy as high as 7 eV that would correspond to much higher temperatures (greater than approximately 1200°C).

4.3 Fast Neutron Energies

Figure 8 shows schematically the neutron flux spectrum as a function of neutron energy in a thermal reactor. Fast neutron energies in thermal reactors may range from 1 keV (corresponding to 10^3 for T_n in Figure 8) to 10 MeV (corresponding to 10^7 for T_n in Figure 8). This exceeds the formation energies about 1 – 10 eV of single defect discussed in Section 4.1. Collective damage is also illustrated in Figure 8 as "Damage function." According to Simmons (1965), neutrons with energies greater than 100 keV (corresponding to 10^5 for T_n in Figure 8) are responsible for most of the damage.



Note: Damage to Reactor Graphite is Illustrated as "Damage function" Shown in Dashed Line (Simmons, 1965, *Radiation Damage in Graphite*).

Figure 8. Schematic Neutron Energy Distributions

4.4 Annealing Parameters

In this section, important parameters relevant to energy release through annealing are summarized based on the review. These parameters are largely empirical and were determined experimentally and/or obtained through fitting Boltzmann relation (e.g., Equation 1).

- *Irradiation temperature* -This parameter refers to the temperature at which a graphite moderator experienced during reactor operations. The lower the temperatures, the higher the stored energies are. A graphite irradiated at or below 250°C (e.g., IAEA-TECDOC-1790) is widely regarded as low irradiation temperature for graphite moderators.

- *Activation energy E and annealing temperature T*- The two parameters are related in Boltzmann equation such as Equation 1. In an experiment that records release rate of stored energy, the activation energy can be determined for a given annealing temperature or vice versa. Both parameters can be related to "migration energy" mentioned in Section 4.2. Some researchers (e.g., Minshall and Jones, 2019, "Modeling the release of Wigner energy during irradiated graphite disposal: are we doing it right?") compared the annealing temperature with the disposal facility temperature to determine if annealing is needed prior to disposal. Note that because fast neutron energy spectrum and variety of defect formation energies, the activation or migration energy is not a singular value but a range of values, or spectrum.

- *Initiation temperature of self-induced Wigner energy release* T_{init} -This parameter refers to the temperature at which a graphite begins to Wigner energy release when heating stops under adiabatic conditions. In the example of Figure 3, the initiation temperature would be less than 120°C for sample A and would be as high as 160°C for sample C.

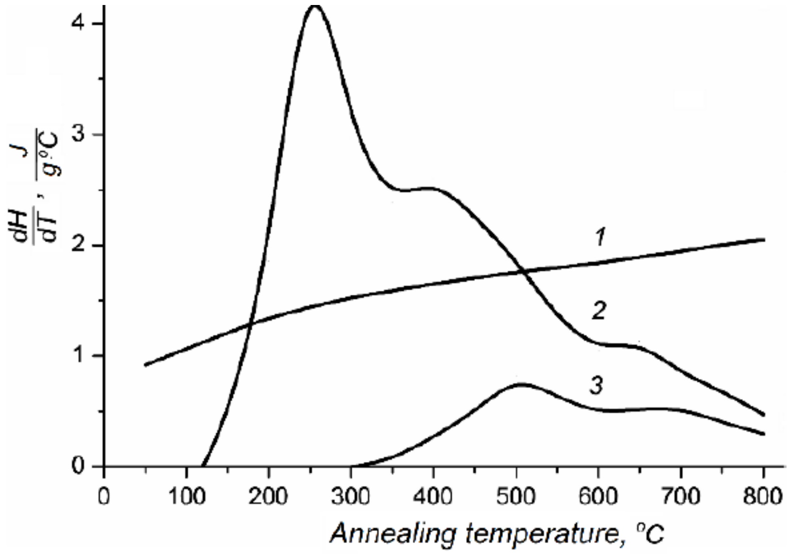
- *Energy release rate as a function of increasing temperature*, or dH/dT (where H is energy released in J and T is annealing temperature in °C) – This is a physical quantity that has the same unit of material specific heat $C(T)$, J/g-°C. In an adiabatic heating condition, only if the following condition is met, the self-sustaining release of stored energy for adiabatic conditions is possible:

$$\frac{dH}{dT} > C(T) \quad \textbf{(Eq. 2)}$$

Figure 9 shows an example of the relationship between dH/dT (curves 2 and 3) of a graphite sample subjected to adiabatic heating and the specific heat of this sample (curve 1). Apparently, according to the researchers Bespala et al., 2015, "Analysis of Wigner Energy Release Process in Graphite Stack of Shut-Down Uranium-Graphite Reactor," the location of curve 3 would not be able to sustain releasing the stored energy when the heating stops. The location of curve 2 would be able to sustain releasing the stored energy when the heating stopped at about 175°C. The temperature increases after 175°C was caused by the released energy until about 500°C.

In the example shown in Figure 9, for the curve 2, the initiation temperature of self-induced Wigner energy release is 175°C, which demonstrates the relationship between the two parameters: dH/dT and T_{init}.

The "cooler region" refers to a region of a graphite that has relatively lower temperature compared to other regions of a graphite moderator block. The cooler regions have higher stored energies. Details depend on specific reactor and graphite moderator designs. Quantitative information must be obtained through experiment or numerical heat transfer simulation (e.g., Pavliuk et al., 2019) of a specific graphite sample.



Note: Sample: 1 – the Specific Heat of the Graphite Sample; 2 and 3 are the Wigner Energy Release Rates as a Function of Annealing Temperature Measured at Different Locations of the Graphite Sample (Bespala et al., 2015, "Analysis of Wigner Energy Release Process in Graphite Stack of Shut-Down Uranium-Graphite Reactor").

Figure 9. The Spectrum of Wigner Energy Release

5 Safety Considerations in Managing Irradiated Graphite Waste

Like any radioactive waste, the ultimate fate of irradiation graphite waste is proper disposal underground or equivalent "safe house." Safety concerns on pre-disposal management and post-closure of disposal facilities arose upon decommissioning of reactors containing the irradiated graphite. By 2006, the international experts appeared to reach the consensus that the irradiated graphite would not self-ignite during all pre- and post-disposal stages (IAEA-TECDOC-1521, *Characterization, Treatment and Conditioning of Radioactive Graphite from Decommissioning of Nuclear Reactors*). The issue of stored energy, however, does not go away from safety concerns. Another concern in managing irradiated graphite waste is the release of radionuclides, especially the gaseous form like hydrogen and carbon-14.

5.1 Predisposal Management

Graphite waste management strategies vary from countries to countries. In this report, the United Kingdom (UK) experience is particularly reviewed, especially their approach to managing Windscale Pile graphite waste. In R&D Technical Report P3-80/TR, options of treating irradiated graphite prior to storage and final disposal were proposed including the following:

- Packaging (with or without cement)
- Annealing
- Incineration
- Steam reforming (pyrolysis)
- "Do-nothing"

Options of vitrification and recycling were also proposed (EPRI, 2006) but are not covered in this review since little attention was drawn. R&D Technical Report P3-80/TR does not conclude which option is more advantageous than the other citing lack of sufficient information. The years followed, some studies on incineration and steam reforming were reported in IAEA-TECDOC-1521. Nevertheless, these reports appeared to generate more questions than solutions in licensing and operation in addition to concerns on gaseous radionuclides and CO_2 as a greenhouse gas release into atmosphere (EPRI, 2006; IAEA-TECDOC-1521). IAEA-TECDOC-1521 does not endorse one or the other. The report recognized that Wigner energy is an issue for reactors operated at low temperatures including Hanford Site reactors with comments:

> The potential risk of triggering an inadvertent release of Wigner energy in these reactors while handling and processing individual graphite blocks during decommissioning, along with the potential for releasing energy during any storage period, packaging, conditioning, and even in final waste repository, is small but requires assessing.

At a 2007 conference reported in IAEA-TECDOC-1647, a UK conceptual plan was presented that Windscale Piles 1 and 2 graphite waste would be packaged and stored without annealing. The conference attendees expressed concerns with this plan. They also recognized lack of data and uncertainties in data collection for supporting annealing. About the same time, a UK NDA, 2007, *Packaging of Windscale Pile Reactors Core Graphite and Aluminum Charge Pans (Conceptual Stage) – Summary of Assessment Report*, document indicated that packaging is for interim storage and further decisions are dependent on demonstrations of (1) that storage and disposal without annealing is passively safe or (2) that annealing can be carried out successfully. Up until now, the UK NDA's baseline strategy is same as 2014 (UK NDA, 2014, "*Higher Activity Waste – Strategic Position Paper on the Management of Waste Graphite*"):

> …to dismantle reactor cores following a period of quiescence (typically 85 years) and package the graphite for disposal.

Geological disposal facility is considered in England and Wales while near-surface disposal facility is considered in Scotland.

It is worthwhile to notice that in both IAEA-TECDOC-1790 and the 2019 international conference on graphite waste (Wickham et al., 2019, "Irradiated Graphite Processing Approach: Update on Project 'GRAPA'"), Brookhaven Graphite Research Reactor (BGRR) decommissioning experience generated a positive response (IAEA-TECDOC-1790). In this approach, the graphite was dismantled without annealing. The graphite was dismantled in "breaking and shoveling" manner without incidents.

5.2 Annealing or Not?

Early understanding on managing the graphite irradiated at low temperature was to anneal the stored energy by controlled thermal heating. Efforts have been focused on quantifying activation energy distributions, i.e., fitting Boltzmann distribution parameters to experimental measurement data (e.g., R&D Technical Report P3-80/TR; BNL-77205-2006-IR; ORNL/TM-2011/378; Guppy et al., 1999; Minshall and Wickham, 1999). There are also numerical simulation studies based on experimental data to predict the release rate of stored energies during annealing processes (e.g., Dostov, 2005, "A Method of Calculating the Rate of Release of Wigner Energy in Heat Conduction Problems for Irradiated Graphite"). For graphite within reactors during operation, it is believed that the unplanned release of stored energy was successfully controlled by regular annealing (IAEA-TECDOC-1790). For irradiated graphite waste management, annealing is not a widely adopted strategy to release the stored energy but has been continually studied. Because of large variation in irradiation conditions, the experimentally measured data or simulation based on the measured data, in general, and lack of a wide range of applications.

The latest understanding is similar to early studies. That is, the activation energies for defect migration follows Boltzmann distribution (e.g., Equation 1) and the distribution parameters must be determined by fitting the measured data (Minshall and Jones, 2019). No theories or models can provide a precise activation energy distribution for a given irradiated graphite. The difficulty lies in a large range of uncertainties in experimental data analysis results, as well as in data analysis methods (Minshall and Jones, 2019), which complicates annealing decision making and process design.

Since 2012, when dismantling of BGRR's graphite during decommissioning was completed, some countries are adopting the BGRR approach (e.g., Dragolici, 2019, "Horia Hulubei National Institute of R&D for Physics and Nuclear Engineering IFIN-HH"). UK's strategy to Windscale Piles 1 and 2 is on one hand packaging and storage and on the other hand continue to fund annealing R&D (UK NDA, 2014; Minshall and Jones, 2019).

Among the suggested energy releasing options, incineration was successfully used to release Wigner energy for small inventory of the irradiated graphite (Vanherck et al., 2019, "Treatment of Thetis Reactor Graphite at Belgoprocess"). The method is not suitable for countries with large quantity of inventory. Other methods are in experimental stages.

5.3 Disposal

Safety considerations in disposal of irradiated graphite waste have been focused on the factors that have potential to trigger sudden release of stored energy in deep geological repository if *the graphite is not annealed prior to disposal*. The concern is, as mentioned previously, inadvertent heating may raise the graphite waste to initiation temperature T_{init}, what may trigger unexpected, self-sustained energy release to further heat up the graphite waste such as shown in Figure 3. To assess this risk, the method of FEPs analysis has been used to identify factors, or FEPs (R&D Technical Report P3-80/TR) that could trigger self-sustained energy release. R&D Technical Report P3-80/TR considered the following FEPs with likelihood of triggering unexpected energy release:

- *Disposal system temperatures* – if the disposal temperature is greater than initiation temperature T_{init}, some stored energy may be released. For example, in UK where the repository temperature is set no greater than 80°C (R&D Technical Report P3-80/TR) and the scientists use this temperature for deciding if annealing is needed or not. The difficulty, however, lies in the uncertainty in determining the initiation temperature of the graphite (Minshall and Jones, 2019).

- *Heat released from cement curing* – some engineered barriers are cementitious materials. If the cement is freshly poured into the repository, the curing process may generate heat that would heat up the graphite. This process, however, is short-lived, estimated to reach up to 60°C in about 100 days and subsequently decline to ambient temperature in about 27 years (Guppy et al., 1999).

- *Metal corrosion and organic waste reactions with microbes* – such exothermic reactions could generate heat that heats up the graphite (R&D Technical Report P3-80/TR) as well as gas generated may stress the graphite. As mentioned in Section B3.2.3, stresses tend to stabilize the interstitials between two lattice planes, a process associated with energy release. This type of reactions, however, is highly dependent on the depth of the repository, types of wastes (containing metal and/or organic materials), disposal site subsurface conditions, groundwater compositions, and the container lifetimes that would allow groundwater flow into waste packages and come in contact with the waste. The reaction rates are generally low compared to heat transfer rates by groundwater flow, which inhibits temperature accumulation.

- *Nuclear criticality* – mostly relevant to disposal with spent nuclear fuel. Criticality can be prevented by employing neutron absorbing materials in waste packages or disposal units containing the graphite. Criticality concern is not just for disposing graphite and is covered in the safety assessment of a disposal program.
- *Human intrusion* – intruder drilling through the graphite was suspected to deliver sufficient energy to graphite to trigger Wigner energy release through stresses. Such an event depends on depths of disposal facilities and engineered barriers for packages containing the graphite waste. The probability of intruder drilling into graphite is low for deep geological disposal. The amount of graphite that would be drilled in an intrusion event is relatively small. In other words, energy produced through drilling would be too low to trigger stored energy release in large scale.
- *Low-probability FEPs external to disposal systems* – such as earthquakes, magma intrusion, and meteorite impact. FEPs like seismic and magma events are generally screened off from the disposal system through site selection. Furthermore, the consequences on radiological safety given rise by these FEPs would probably overwhelm the consequence by the graphite's Wigner energy release alone.

Based on the above identified FEPs, R&D Technical Report P3-80/TR further assessed the impact of temperature rise due to Wigner energy release on performance of a deep geological repository. The impacts include the following:

- Engineered barrier integrity that affects radionuclides with half-life up to 30 years
- Long-lived radionuclide concentrations in groundwater due to altered chemical properties
- Groundwater flow field during the thermal transient
- Long-term groundwater flow field
- Gas migration from the repository

The results concluded that the risk is low if the repository temperature is raised to 78°C and the risk can be high if the repository temperature is brought to 200°C, especially when temperature rise rate is high. In IAEA-TECDOC-1790 report, it was generally accepted that Wigner energy would have low risk to repository.

6 Summary and Conclusions

This report is based on review of past and recent studies on Wigner energy effect in low-temperature irradiated graphite. The review can be summarized as follows:

- Wigner energy may be significant in low-temperature irradiated graphite, especially in cooler regions.
- Total amount of Wigner energy in an irradiated graphite depends on neutron fluences and temperatures during irradiation in reactors.
- Wigner energy can be released in an annealing process through heating. The annealing process parameters may be determined experimentally. There is a large uncertainty in annealing parameters due to both intrinsic, or aleatory, uncertainty and experimental data analysis result, or epistemic, uncertainty. The aleatory uncertainty is intrinsic to irradiation graphite that arises from a variety of defects and their configurations formed during irradiation. The epistemic uncertainty is caused by experimental data analysis methods.
- Windscale incident was not caused by Wigner energy release from the irradiated graphite nor graphite burning.

- For low-temperature irradiated graphite, there is a large range of defect migration energies, corresponding to annealing temperatures ranging from room temperatures to greater than 1200°C.
- Heating the irradiated graphite under adiabatic (a necessary but not sufficient) conditions may initiate self-sustained energy release.
- The impact of low-temperature irradiated graphite on disposal safety is low even without predisposal annealing.
- The UK's baseline strategy to managing Windscale Piles 1 and 2 graphite is packaging and storage before disposal in either geological or near-surface facilities.
- Several countries are following suit of US BGRR in decommissioning reactors contain irradiated graphite.
- R&D in annealing is still underway in both experimental and experimentally supported numerical simulations.

Based on the above summary, it can be concluded that Wigner energy does not present major challenges to disposal of low-temperature irradiated graphite waste without annealing. The risk to disposal facilities brought by irradiated graphite arises from temperature rise due to Wigner energy release from the graphite. However, Wigner energy release can only be triggered by heating. In the absence of heat sources such as waste decay heat, the risk is extremely low. For the same reason, the likelihood of self-sustained energy release from irradiated graphite under adiabatic condition is trivial in the storage and disposal systems without heat sources.

Annealing would not completely release all the Wigner energy in the graphite. Furthermore, annealing of irradiated graphite waste has yet to be demonstrated in terms of safety and cost-effectiveness. Following BGRR's successful dismantling of the graphite waste, some countries are adopting similar approaches.

The current Hanford reactor irradiated graphite waste has been separated from spent fuel, which means that the long-term heat source is absent. Care should be exercised if grouting the waste package containing the irradiated graphite even though the heat released by cement curing is insignificant to trigger sudden release of Wigner energy. Prevention of unexpected Wigner energy release during storage can be further ensured to maintain passive cooling in the facility. Finally, for near-surface disposal, impact of drilling through irradiated graphite is likely low and will be included in safety/performance assessment before the reactor blocks are disposed.

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