Safety Assessment on the Storage of Irradiated Graphite Waste Produced from the Decommissioning of KRR-2

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ABSTRACT

Irradiated graphite waste arising from the decommissioning of KRR-2 (Korean Research Reactor 2) has shown evidence of Wigner energy, specific radioactivity and the resulting radioactive chemicals/ nuclides upon examination of its graphite structure. Plans of annealing and disposal of the irradiated graphite must consider whether the stored Wigner energy will allow for its safe storage. Wigner energy is a latent explosive energy in the graphite structure due to fast neutron irradiation that caused carbon atomic rearrangements or dimensional distortions in the graphite crystal lattice. It is important to forecast the thermal stability of this graphite in long-term storage since in potential accident scenarios where high temperatures could occur, the Wigner energy has a greater potential to be released. Since nuclide emissions will also result if the thermal stability of the graphite in storage is not maintained, the elimination or control of Wigner energy content at a lower level in the waste program of the irradiated graphite will be necessary. A fire accident scenario was simulated to assess the feasibility of drum disposal storage of the graphite without any physical and/or chemical treatment of Wigner energy. Simulation results indicate that within 30 minutes of outer drum surface fire exposure, graphite waste containing Wigner energy reaches temperatures sufficient for the graphite to ignite. Therefore it is necessary to remove Wigner energy from the graphite waste for its safe storage.

INTRODUCTION

Graphite has been used as a moderator and reflector of neutrons in more than 100 nuclear power plants as well as many experimental reactors and plutonium production reactors in various countries. Most of the older graphite moderated reactors are already shut down and are awaiting decommissioning planning and preparation. Radioactive graphite dismantling, handling, conditioning and disposal are a common part of the decommissioning activities.

The graphite in some of the experimental and plutonium production low temperature reactors contains a considerable amount of stored Wigner energy. Unexpected release of Wigner energy, mainly in the older graphite moderated reactors, particularly those built to produce plutonium, has caused several incidents, also connected sometimes with fuel failure. The potential risk associated with accumulated Wigner energy is one of the main safety risks to address during graphite waste processing and disposal.

The decommissioning of the research reactors (KRR-1&2) in Korea, which started in 1997, generated 13 tons of radioactive graphite waste [1]. KRR-1, the first research reactor in Korea (TRIGA Mark-II), has been operated since 1962, and KRR-2, the second one (TRIGA Mark-III), since 1972. These two reactors were phased out in 1995 upon reaching their planned operation lifetime as the new and more powerful research reactor, HANARO (High-flux Advanced

Neutron Application Reactor) began operation at the site of the Korea Atomic Energy Research Institute (KAERI) in Daejeon. The KRRs are TRIGA pool type reactors in which the cores are small self-contained units located in tanks filled with cooling water. The KRR-1 is a TRIGA Mark II, which began operation in May of 1962 and could operate at a level of up to 250 kW. The second one, the KRR-2 is a TRIGA Mark III, which could operate at a level of up to 2,000 kW.

Irradiated graphite arising from the decommissioning of KRR-2 (Korean Research Reactor 2) shows evidence of Wigner energy, radioactivity and the resulting radioactive chemicals/nuclides from the study of its graphite structure [2]. Plans of annealing and disposal of the irradiated graphite must consider whether the stored Wigner energy will allow for its safe storage[3]. Wigner energy is a latent explosive energy in the graphite structure due to fast neutron irradiation that caused carbon atomic rearrangements or dimensional distortions in the graphite crystal lattice. It is important to forecast the thermal stability of the graphite in long-term storage since in potential accident scenarios where high temperatures could occur, the Wigner energy has a greater potential to be released [4]. Since nuclide emissions will also result if the thermal stability of the graphite in storage is not maintained, the elimination or control of Wigner energy content at a lower level in the waste program of the irradiated graphite will be necessary.

In this study a fire accident scenario was simulated involving irradiated graphite that is planned for disposal in a radioactive waste storage facility. The purpose of the simulation was to evaluate the thermal stability of graphite by establishing whether or not the Wigner energy of the irradiated graphite affects its storage stability when involved in a sudden fire accident.

WIGNER ENERGY

Fast neutron irradiation of graphite in a nuclear installation displaces carbon atoms from their normal positions in the graphite lattice, creating a variety of defects through combinations of these displaced atoms. When displaced atoms return to their original state by recombining with vacancies within the lattice, a process also known as annealing, there is a release of the Wigner energy in the form of heat.

In graphite irradiated at room temperature, very large levels of stored energy can accumulate, values of up to 2,700 J/g have been recorded [5]. If all this energy were released as heat it would lead to a temperature rise of approximately 1,500 °C. However, in air, the graphite would start to thermally oxidize before this temperature rise is attained.

Most of the reported data shows high Wigner energy content and high deformations of the various physical properties of the irradiated graphite according to the neutron dose history, e.g., $E > 50$ keV and $T > 300°$ C, in general [6]. In spite of the peculiar characterization of the KRR-2 graphite affected by low neutron flux (short operating time) at low temperature ($T < 100°$ C), the release of Wigner energy from the graphite was still observed.

The release of Wigner energy from graphite in any environment subsequent to its irradiation is dependent upon the spectrum of defects created by irradiation. In general, the higher the original irradiation temperature, the higher the temperature of any subsequent environment required to achieve a given rate of annealing and thus rate of Wigner energy release.

The total amount of stored energy in any given graphite is generally not a useful indicator of graphite behavior. To anneal all defects in the lattice of a highly irradiated graphite sample within a practical period of time, and thus release all its Wigner energy, temperatures of up to 2000° C would be required. At the temperatures more likely to be experienced by the graphite in practice, only a fraction of the total stored energy could be released over a reasonable timescale [7].

WIGNER ENERGY RELEASE MODEL

There has not been any detailed attempt to give a theory of the release of stored energy in terms of the defect concentrations, although correlation of total stored energy with simple models has been achieved. The importance of stored energy release in a variety of reactor accidents has led to detailed empirical methods of treatment of the release applicable to arbitrary temperature-time relationships.

Simmons[8] has proposed a very general expression for the energy release rate with respect to time:

$$\frac{dS}{dt} = f(S)e^{-\frac{E}{kT}} \qquad \text{(Eq. 1)}$$

where S is the energy released or remaining, t is the time, E is the activation energy, k is the Boltzmann constant, and T is the temperature.

A single activation energy model is unlikely to be correct, given the very wide range of measurement temperatures at which energy release is observed when the temperature is raised at a constant rate. A simple model in which the activation energy varies was devised by Vand and further developed by Primak [4]. In the simplest form, due to Vand, it is assumed that the energy release process for each group of defects obeys first order kinetics. Thus for a group with activation energy E at constant temperature T

$$\frac{dS}{dt}(E,t) = -\nu\, S(E,t)\, e^{-\frac{E}{kT}} \qquad \text{(Eq. 2)}$$

where ν is a constant frequency factor. In an isothermal anneal Eq. (2) integrates to

$$S(E,t) = S_0(E)e^{-\nu t \exp\left|-\frac{E}{kT}\right|} \qquad \text{(Eq. 3)}$$

where the initial stored energy S_0 is given by

$$S_0 = \sum_i S_0(E_i)\ . \qquad \text{(Eq. 4)}$$

From Eqs (2) and (3), the parameters of the frequency factor ν and the activation energy E are the key factors to be calculated for modeling the thermal recovery process (model for annealing) of the KRR-2 graphite.

Bridge and Mottershead[9] found to a good approximation

$$E = (33.7 - 1.83\log a)T \times 10^{-4} - 0.037 \ \text{ eV} \qquad \text{(Eq. 5)}$$

with a in °C /min, valid in the range $0.1 < a < 2 \times 10^3$ °C /min.

From the experiment results, the frequency factor for the KRR-2 graphite is 4.66×10^{14} which is larger than Bridge’s frequency factor 7.5×10^{13}.

As shown in Fig. 1 the derived energy release model is good for the irradiated graphite arising from KRR-2. Although the small deviation occurred at a peak point (200° C ~250° C) and a tailing point (330° C ~500° C).

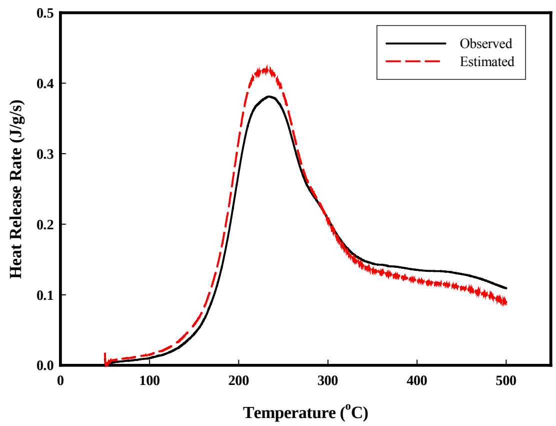


Fig. 1. Heat release curve using variable activation energy model

STORAGE AND ACCIDENT SCENARIO

The irradiated graphite waste arising from the decommissioning of KRR-2 is expected to be packaged in 200-liter drums. The graphite waste then will be temporarily stored in the KRR-2 reactor hall until a low- and intermediate-level radioactive waste disposal site is operational. For the purposes of modeling, it has been assumed that the graphite waste would be suitable for packaging in a non-encapsulated from, and therefore a heat generating binder would not be used.

The thermal behavior of a 200-liter drum exposed to a 1000˚ 1-hour fire was modeled. To simulate the fire, a radiant heat flux was applied to the outer surface of the drum. After the period of heating, the drum was allowed to cool down in ambient temperature air. This cooling was accomplished in the model by placing a heat sink, at a constant temperature of 25 on the outer surface of the drum. No heat inputs other than those arising from Wigner energy were applied.

MODELING FOR SAFETY ASSESSMENT

For modeling purposes it was assumed that the graphite waste would be packaged as a powder in a 200-liter drum and that the Wigner energy would be uniformly distributed in the packaged graphite waste. The key material properties are summarized in Table I. It was assumed that these properties would not vary significantly with temperature or time.

Table I. Material Properties of Graphite and Drum

Material	Heat output	Density (kg/m^3)	Thermal conductivity (W/m/K)	Specific heat capacity (J/kg/K)
Graphite	Wigner energy	1600	2.1	1300
Drum (steel)	0	7800	15	1006

The drum was modeled using a finite element analysis technique with commercially available software ABAQUS. Since the drum has cylindrical symmetry, it was necessary to model only a quarter of the drum as illustrated in Fig. 2.

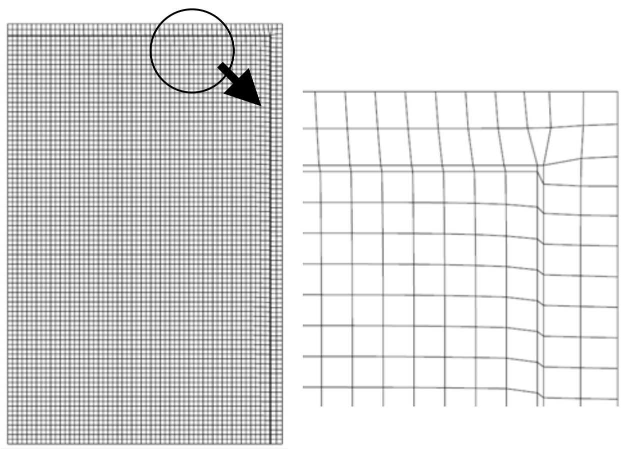
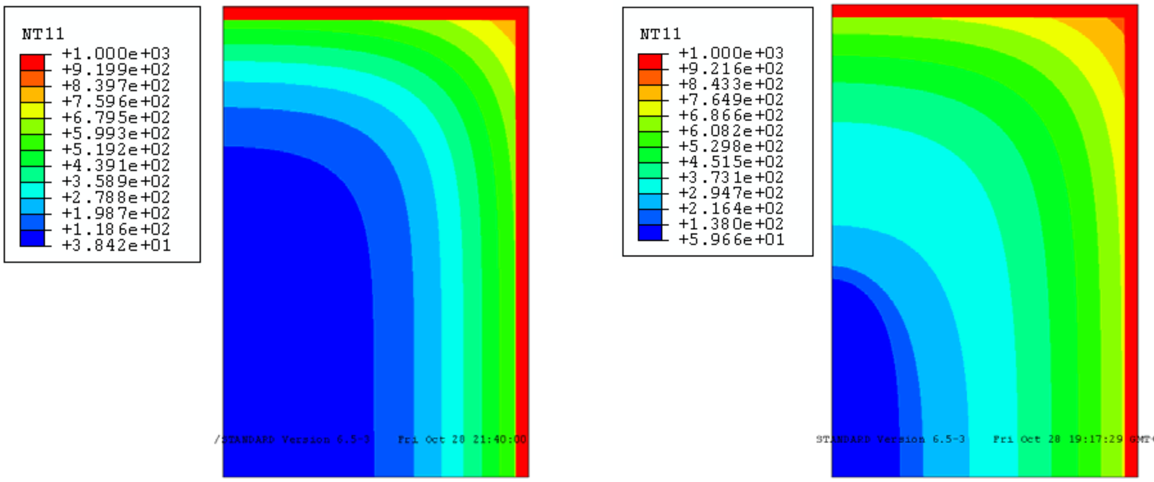


Fig. 2. The modeled drum for safety assessment

The calculation of Wigner energy release from the graphite was performed by a user subroutine employing the variable activation energy release model. The Wigner energy released was calculated on an element-by-element basis, and applied as a heat source within each element.

RESULTS AND DISCUSSION

Fig. 3 shows temperature profiles in the graphite waste in the drum after the 1-hour fire. The heat from the fire penetrates the drum and raises the temperature of the edge of the waste to a peak of about 800˚ with no Wigner energy and about 900˚ with Wigner energy, respectively. However, the graphite at the center only displayed a small temperature rise during the simulated accident due to the low thermal conductivity of the irradiated graphite modeled. Analysis of heat distribution within the graphite showed that the elevated temperatures encompassed approximately 50% of the waste. However, the limited amount of data in the variable activation energy model affected this result.



(a) no Wigner energy (b) with Wigner energy

Fig. 3. Temperature profile of the graphite after fire accident

The temperature rise at the outer surface of graphite waste during the fire accident scenario is shown in Fig. 4. Within 2000 seconds the outer surface of the irradiated graphite with Wigner energy reaches 500˚ which is the starting temperature for the thermal decomposition of graphite [10]. From these results it is anticipated that within 30 minutes, graphite waste near the drum surface containing Wigner energy reaches sufficient temperature for the graphite to ignite. Therefore it is necessary to remove Wigner energy from the graphite waste for its safe storage.

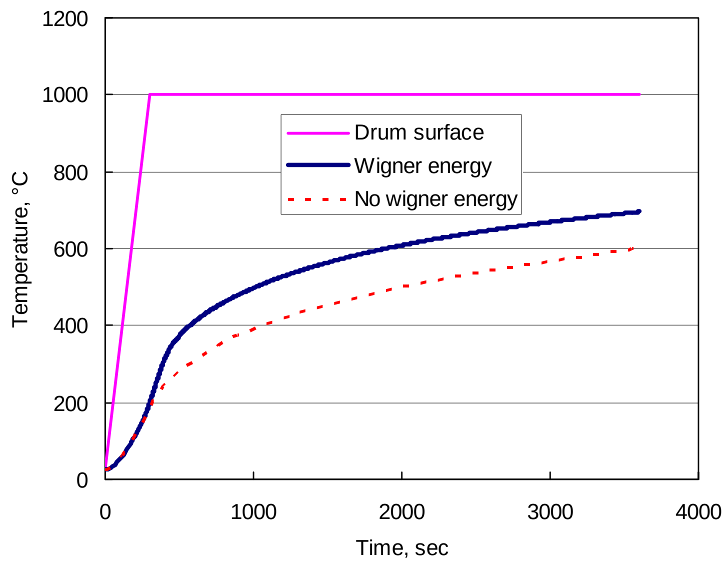


Fig. 4. Temperature rise at outer surface of graphite

CONCLUSION

A model for the release of Winger energy from irradiated graphite has been written into a computer subroutine and applied with a finite element modeling technique to assess the safe storage of waste packages containing decommissioned graphite from KRR-2. Modeling of a fire accident for the irradiated graphite from KRR-2 was performed.

In this study a fire accident scenario was simulated involving irradiated graphite that is planned for disposal in a radioactive waste storage facility. The purpose of the simulation was to evaluate the thermal stability of graphite by establishing whether or not the Wigner energy of the irradiated graphite affects its storage stability when involved in a sudden fire accident. Simulation results indicate that the exposure of the graphite waste to a severe fire accident can result in the rapid release of Wigner energy. This response could cause a large temperature rise, possibly resulting in graphite ignition. Therefore it is necessary to remove Wigner energy from the graphite waste for its safe storage.

ACKNOWLEDGEMENTS

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