RADIOLOGICAL AND PHYSICAL CHARACTERIZATION OF THE IRRADIATED GRAPHITE WASTE ARISING FROM THE DECOMMISSIONING OF KOREAN RESEARCH REACTOR

D. G. Lee, K. H. Chung, K. W. Lee, K. J. Jung, W. Z. Oh
Korea Atomic Energy Research Institute

ABSTRACT

Radioactive graphite dismantling, handling, conditioning and disposal are common parts of the decommissioning activities. At the Korean Research Reactor 2 (KRR-2), nuclear graphite was used as a moderator in thermal column, which was used to irradiate experimental specimens. The radiological and physical properties of graphite waste from the decommissioning of KRR-2 have been investigated to analyze their irradiation effect. It was found that the various radionuclides are present - mainly H-3, C-14, Co-60, Cs-134, Eu-152 and Eu-154. The widely known effect of physical properties change in irradiated graphite was observed in the experiment. The basic theories of the accumulation of stored energy (Wigner energy) in graphite waste and its release are reviewed. In order to investigate the release of Wigner energy activation energy spectra were derived from differential scanning calorimetry of samples of the KRR-2 graphite. The graphite waste retain substantial amounts of Wigner energy and thus to support their packaging, storage and ultimate disposal, it is necessary to induce the release of the Wigner energy. The graphite waste will be heated in annealing ovens, which results in the release of significant amounts of Wigner energy.

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 therefore decommissioning planning and preparation represents a very serious challenge. Radioactive graphite dismantling, handling, conditioning and disposal are a common part of the decommissioning activities.

The radioactive graphite waste has different characteristics compared to other radioactive waste due to its physical and chemical properties. It includes various radionuclides such as tritium and C-14, as well as corrosion/activation products (Co-57, Co-60; Mn-54; Ni-59; Ni-63; Na-22), fission products (Cs-134, C-137; Sr-90; Eu-152, Ce-144) and small amount of uranium and transmutation elements (Pu-238, Pu-239; Am-241, Am-243) [1].

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, caused several incidents, also connected sometimes with fuel failure. Potential risk connected with accumulated Wigner energy is one of the main factors which has to be taken into account during graphite waste processing and disposal.

The decommissioning of the research reactors (KRR-1&2) in Korea, which started in 1997, generated 13 tones of radioactive graphite waste [2]. 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. The operation of both of them was phased out in 1995 due to reaching their lifetime and operation of the new and more powerful research reactor, HANARO (High-flux Advanced Neutron Application Reactor) at the site of the Korea Atomic Energy Research Institute (KAERI) in Daejeon. Both are TRIGA pool type reactors in which the cores are small self-contained units sitting in tanks filled with cooling water. The KRR-1 is a TRIGA Mark II, which went through its first criticality 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.

The decommissioning project of these two research reactors was started in January 1997 and will be completed by 2008. The aim of the decommissioning activities is to decommission the KRR-1&2 and to decontaminate the residual building structures and the site to release them as unrestricted areas. According to the schedule, the practical decommissioning activities were started in June 2001 by cleaning first the radioisotope production equipment and experimental laboratories in the KRR-2. Cleaning of more seriously contaminated areas such as the lead hot-cell and concrete hot-cell, then followed. The reactor of KRR-2 is being dismantled starting in 2003.

In this study, the radiological and physical properties of graphite waste from the decommissioning of KRR-2 have been investigated to analyze their irradiation effect. The basic theories of the accumulation of stored energy (Wigner energy) in graphite waste and its release were reviewed. In order to investigate the release of Wigner energy activation energy spectra were derived from differential scanning calorimetry of samples of the KRR-2 graphite.

GRAPHITE SAMPLES

Sample graphite material was obtained from the graphite blocks used in thermal column of KRR-2. The analyzed compositions of graphite sample are shown in Table I. The sample graphite was found to contain over 99.99% carbon, 190 ppm chlorine and about 20 ppm ash.

Table I. The compositions of sample nuclear graphite (dry weight basis)

<table>
<tr><td rowspan="2">Proximate analysis</td><td colspan="2">Volatile matter</td><td colspan="2">Fixed carbon</td><td>Ash</td></tr>
<tr><td colspan="2">2.2%</td><td colspan="2">99.8%</td><td><500 ppm</td></tr>
<tr><td rowspan="2">Ultimate analysis</td><td>C</td><td>H</td><td>O</td><td>N</td><td>Cl</td></tr>
<tr><td>>99.98%</td><td><0.001%</td><td><0.001%</td><td>-</td><td>190 ppm</td></tr>
</table>

RADIOLOGICAL CHARACTERISTICS OF THE IRRADIATED GRAPHITE FROM KRR-2

Determination of Gamma Nuclides

To obtain radionuclide inventories (gamma-emitters) in the irradiated graphite, MCA (multi-channel analyzer) was used. From the results of MCA analysis, it was found that various radionuclides (mainly Co-60, Ba-133, Cs-134, Cs-137, Eu-152 and Eu-154) are present in the irradiated graphite from KRR-2. The radioactivity of the graphite samples from various positions of the thermal column of KRR-2 is summarized in Table II.

Table II. Radioactivity of the irradiated graphite from KRR-2

Sample No.	Position	Surface Dose Rate (mSv/hr)	Specific Activity (Bq/g)
11	1M11	0.17	1049.35
45	1J01	0.22	4157.38
48	1J04	0.32	5252.24
53	1J05	1.95	8679.78
56	1J12	0.55	5619.96

Determination of Beta Nuclides

The method used in measurements of H-3 and C-14 contents in the graphite samples was presented in Ref.[3]. The method is based upon chemical separation of radionuclides from the graphite sample being assayed in gaseous form with subsequent transition into liquid phase and measurement of activity by means of liquid scintillator. The specific activity of H-3 and C-14 in the irradiated graphite of KRR-2 was evaluated as 102.57 Bq/g and 114.34 Bq/g, respectively.

Radionuclide C-14 is generated in the graphite due to two main neutron reactions: $^{13}C(n,\gamma)\,^{14}C$ and $^{14}N(n,p)\,^{14}C$. Nitrogen in graphite is in bound state and as a gas filling up the graphite pores. Porosity of the graphite is about 26%. Contributions of these two neutron reactions to build-up of C-14 in graphite were evaluated, and the evaluation revealed that neutron reaction with nitrogen gave a major contribution.

Main contribution to accumulation of tritium in graphite is given by neutron reactions with Li-6 and N-14 nuclei. Generated triton will stay in the graphite matrix with high probability because mean free path of triton is smaller than size of graphite grains and larger than size of pores. However, some fraction of tritons generated near to grain boundaries has no energy high enough to penetrate through pore or gap. Therefore, some fraction of tritium has a possibility to remain in the gas filling up the pores and gaps.

PHYSICAL CHARACTERISTICS OF THE IRRADIATED GRAPHITE FROM KRR-2

Density Change

Irradiation of graphite with energetic neutrons leads to dimensional (density) changes of considerable magnitude which must be accommodated for in reactor design. Graphite may be considered to be an aggregate of crystallites of identical properties. The irradiation behavior of the crystallite is that they grow perpendicular to the basal planes and shrink parallel to the planes.

The density change of the irradiated graphite from KRR-2 was evaluated by using the following equation.

$$Q = \frac{A}{A-B} Q_0$$

where Q : density of solid

Q_0 : density of liquid (distilled water)

A : weight of solid in air

B : weight of solid in liquid (distilled water)

The experiment results are summarized in Table III. As shown in Table III, the density of graphite sample was increased by about 3.5% and the change rate is proportional to the neutron flux (radioactivity).

Table III. The density change of the irradiated graphite from KRR-2

Graphite Sample	Unirradiated	29C	48A
Specific Activity (Bq/g)	-	186.99	1512.13
A (g)	6.9782	8.8895	5.6537
B (g)	2.8503	3.8096	2.4053
Density (g/cm^3)	1.6875	1.7468 (3.51% ↑)	1.7374 (2.96% ↑)

It has been reported that the number of relatively large pores are reduced by neutron irradiation leading to an increase in the density until saturation occurs at a fluence corresponding to the maximum shrinkage [4]. After that the volume increase occurs by an increase in the number of small pores in the graphite.

Stored Energy

Fast neutron irradiation of graphite in nuclear installation displaces carbon atoms from their normal positions in the graphite lattice, creating a variety of types 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 temperature rises in the region of 1,500 °C. However, in air, before this temperature is reached the graphite would start to thermally oxidize.

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 well observed, not a little.

To measure the Wigner energy content of the KRR-2 graphite the heat release rate was observed by DSC (differential scanning calorimeter). The graphite samples were heated from 25 °C to 500 °C with 10 °C/min heating rate. Figure 1 shows a comparison of stored energy (Wigner energy) during linear temperature rise in DSC measurement.

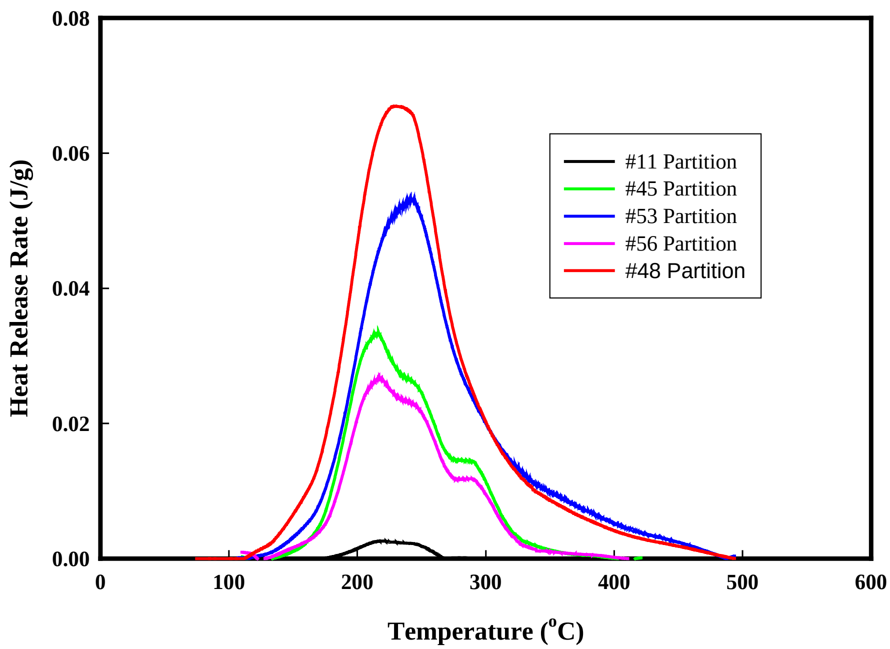

Fig. 1. A comparison of Wigner energy for the irradiated graphite from KRR-2

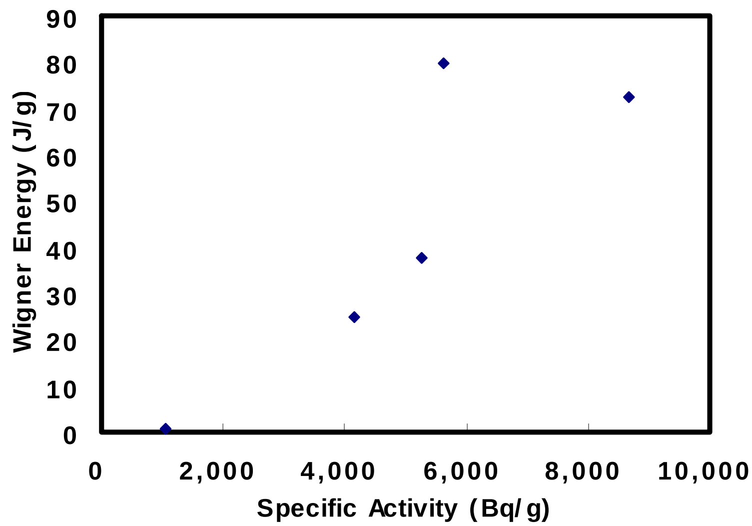

Fig. 2. The relationship between Wigner energy and radioactivity for graphite sample

Graphite samples of the KRR-2 show a typical heat release curve in an elevated heat treatment at an inert gas atmosphere. The stored energy of graphite sample is measured 0.57 ~ 79.8 J/g and the energy release can be started when the heating temperature is raised more than 120 °C. The maximum release temperature is 200 ~ 250 °C. As shown in Figure 2 the Wigner energy is proportional to radioactivity. The most of stored energy can be released when the irradiated graphite from KRR-2 is heated more than 300 °C.

CONCLUSION

The radiological and physical properties of graphite waste from the decommissioning of KRR-2 have been investigated to analyze irradiation effect. It was found that the various radionuclides are present - mainly H-3, C-14, Co-60, Cs-134, Eu-152 and Eu-154. The widely known effect of density change in irradiated graphite was observed in the experiment. The radioactive graphite has different characteristics from the other radioactive waste due to its physical and chemical properties and also because of the presence of tritium and carbon-14. To quantify the stored energy of the irradiated graphite the temperature of the graphite sample was raised in a controlled manner and the energy release rate was measured by DSC (differential scanning calorimeter). It was found that not a little quantity of the stored energy was contained in the irradiated graphite from KRR-2. The irradiated graphite from KRR-2 should be annealed at over 300 °C to remove stored energy.

ACKNOWLEDGEMENTS

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