Wigner Energy Storage and Release Characteristics of the Irradiated Graphite from the Decommissioning of KRR-2

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1. Introduction

Graphite sample of the KRR-2 (Korean Research Reactor 2, TRIGA Mark-III type) shows a typical heat release curve in an elevated heat treatment at an inert gas atmosphere. This heat release phenomenon by heat treatment (annealing) of the irradiated graphite is so called the release of 'Wigner Energy' that is latent or stored as an internal energy in the graphite structure caused by carbon atomic rearrangements or dimensional distortions in the graphite crystal lattices[1]. To quantify the stored energy of the irradiated graphite the temperature of the graphite sample was raised in a controlled way and the energy release rate was measured by DSC (differential scanning calorimeter) [2,3]. To measure the Wigner energy content of the KRR-2 graphite the heat release rate was observed and was modeled by the general annealing equation in this study. But also, to characterize the storage of the Wigner energy in the irradiated graphite the gradient of this internal energy in the graphite blocks due to distance from reactor core was compared.

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℃, in general [4]. In spite of the peculiar characterization of the KRR-2 graphite affected by low neutron flux (short operating time) at low temperature (T < 100℃), the release of Wigner energy from the graphite was well observed, not a little.

2. Method and Results

2.1. Released Energy Measurement

To determine the propagation of the energy release from the irradiated graphite by DSC measurement (DSC Q(100), TA Instruments) two well-known running methods were selected[2,3].

(a) Keep isothermal conditions and observe the sample temperature-time relation in two successive runs.

(b) While constant heating (linear temperature rise method), observe the difference in energy supply.

2.2. Stored Energy Release Model

The energy release is expressed in the general form with respect to time[3]:

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

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 modified equation was proposed in terms of variable activation energy model[2,5].

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

where ν is a constant frequency factor. In an isothermal anneal Eqn. (2) [(a) method] integrates to

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

where the initial stored energy S_0 is given by

$$S_0 = \sum_i S_0(E_i).$$

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

2.3. Results

Fig. 1 shows a comparison of the thermal annealing during a linear temperature rise [constant rate of heat flux, method (b)] in DSC measurements. Three samples were Used: (1) an irradiated graphite, (2) an irradiated graphite after annealing of sample (1), and (3) an unirradiated graphite sample. The irradiated graphite sample shows an exothermic heat transition, which is not much but approached to approximately 60 J/g, at an inert medium (helium) annealing. Sample of the irradiated graphite after annealing comes from sample (1) shows no output of any type of stored Wigner energy, but it shows a simple heat absorption curve during heat processing. This means that the Wigner energy of the original irradiated graphite was already released at the first heat treatment and the residual heat capacity of the sample (2) reveals the original property of the unirradiated graphite, or seems to be, like the result of sample (3). It is not conclusive that the distorted graphite lattice of the irradiated graphite becomes back to the formation of the original unirraidated graphite structure by means of annealing, but the enlarged heat absorption capacity of the irradiated sample (1) shows more than 2 times greater to the unirradiated sample (3), except a sudden heat release periods in DSC measurement.

The activation energy for annealing is ranged between 1.4 and 1.6 eV, for a while under studying

KRR-2 graphite, in which the magnitude is similar to other experimental suggestions [5].

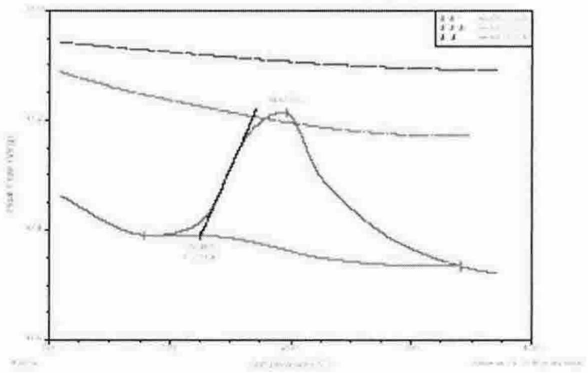

Figure 1. DSC results of the irradiated graphite and unirradiated graphite sample by linear temperature rise method. (1) Irradiated graphite (green line), $\triangle H$=-60 kJ/g, (2) Irradiated graphite after annealing (blue dash line), (3) Unirradiated graphite (red dash dot).

3. Conclusion

Wigner energy of the irradiate graphite from KRR-2 was measured by using DSC. Not a little quantity of the stored energy was contained in it. Conclusively, irradiated graphite shows a build-up heat capacity, i.e., heat absorbing capacity, during heat supplying period, except a sudden heat spouting, in DSC test.

The activation energy of the heat release model of the irradiated graphite was in the range of the reported data.

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