The Release Rate of Wigner (stored) Energy for Irradiated Graphite Reflector

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1. Introduction

In nuclear reactors, graphite is mainly used as neutron reflector or as neutron moderator. Therefore graphite is positioned inside or close to the reactor core and experience high dose of neutron irradiation. An important phenomenon resulting from the neutron damages is the striking release of stored energy at around 200°C after certain buildup of the irradiation. Crystal lattice defects introduced by fast neutron irradiation increase the energy of the graphite crystals. Increasing the temperature above that at which the defects were produced allows the defects to rearrange themselves to states of lower energy, with the excess energy being released as heat. This stored energy is often referred to as Wigner energy. In graphite irradiated at room temperature, very large levels of stored energy can be accumulated, values of up to 2,700 J/g which if released as heat under adiabatic conditions would raise its temperature by ~ 1500°C [1]. Therefore, Wigner energy is the one of the problematic damages in low-temperature and high neutron flux irradiation condition.

However, in safety assessments, it is not the total amount of stored energy that is of prime concern but the release rate of this energy with temperature rise and the temperature at which this release rate becomes significant [2]. This paper will introduce some theory of the release of stored energy in graphite and discuss about safety assessments of research reactor.

2. Theoretical backgrounds and Methods

The simplest way of considering how much a massive amount of heat released as Wigner energy influences, is to define an effective specific heat:

$$C_p^{'} = C_p - \frac{dS}{dT} \qquad (1)$$

If equation(1) is negative over a temperature range, the effective specific heat is negative and the sample is self-heating, and the possibility of large, virtually adiabatic, temperature rises exists[1]. Therefore, the problem is to determine dS/dT. Fig. 1 shows the rate of release of stored energy on different conditions and specific heat of graphite.

The rate of release of stored energy depends on both the temperature and the amount of released energy. In addition, the kinetics of the release of stored energy are influenced by the irradiation conditions (temperature, dose of irradiation and the state of graphite)[3].

Simmons (1965) has proposed a very general expression for the energy release rate with respect to time:

$$\frac{\partial S}{\partial t} = F(s)e^{-\frac{E(S)}{kT}} \qquad (2)$$

where E(S) (eV) is the activation energy as a function of stored energy remaining in the sample, T(K) is the temperature, k is Boltzmann's constant and f(S) is a function which characterizes the state of the sample[2].

There are various models which deal with the kinetics of the release of stored energy [2].

2.1 Variable activation energy model

The function f(S) and E(S) can be derived from linear rise experiments using assumptions about activation energy as a function of temperature and linear rise rate. For example, a pair of linear rise experiments takes at different rates is conducted at each temperature T_1 and T_2. From these experiments, following equation is derived:

$$\frac{E(S)}{k}\left(\frac{1}{T_1} - \frac{1}{T_2}\right) = \ln\left[\left(\frac{dS}{dt}\right)_2\right] - \ln\left[\left(\frac{dS}{dt}\right)_1\right] \qquad (3)$$
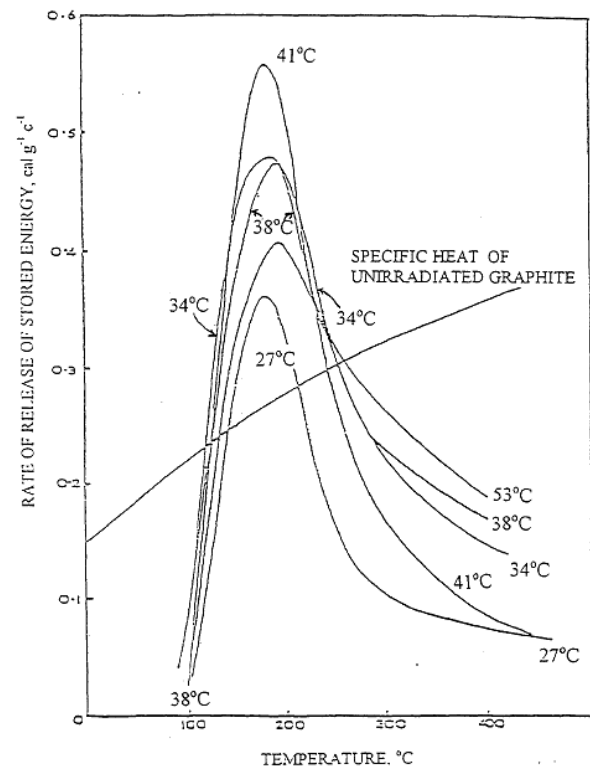
From this, the rate of release can be given as:

Figure 1: Linear Rise Curves for Graphite Irradiation Below 150°C

$$\frac{dS}{dt} = \left(\frac{dS}{dt}\right)_1 \exp\left[-\frac{E(S)}{k}\left(\frac{1}{T}-\frac{1}{T_1}\right)\right] \quad (4)$$

2.2 Constant activation model

A simple approximation can be made by assuming constant activation energy. Thus, the rate of release equation becomes:

$$\frac{dS}{dt} = f(s)e^{-\frac{E}{kT}} \quad (5)$$

2.3 Constant frequency factor model

This model was proposed by Vand and developed later by Primak. 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:

$$\frac{dS}{dt}(E,t) = -\nu S(E,t)e^{-\frac{E}{kT}} \quad (6)$$

where ν is a constant frequency factor.

The calculation will be mainly based on the third one. In an isothermal annealing, equation (6) integrates to [1]

$$S(E,t) = S_0(E)e^{-\nu t \exp\left(-\frac{E}{kT}\right)} \quad (7)$$

An inflection point is located at $E_0 = kT\ln(nt)$ with a value 0.368. The variation with E is so rapid that a step function of unit height located at E_0 can be applied. As a result, equation (7) can be approximated by

$$S(t) = \int_{E_0}^{\infty} S_0(E)dE \quad (8)$$

which gives on differentiation with respect to time

$$\frac{dS}{dt} = -S_0(E_0)\frac{dE_0}{dt} = -\frac{S_0(E_0)kT}{t} \quad (9)$$

with following conditions

$$\left(\frac{E_o}{kT}\right) + \ln\left(\frac{E_o}{kT}\right) = \ln(\nu t), \ \frac{dE_o}{dt} = ka\left(\frac{E_0}{kT}\right)\frac{\left[\frac{E_o}{kT}+2\right]}{\left[\frac{E_o}{kT}+1\right]} = kaU \quad (10)$$

where U is practically constant for a given value of a, that is $E_0 \propto T$. As a result, the activation energy spectrum can be obtained as

$$S_0(E_0) = -\frac{\left(\frac{dS}{dT}\right)_a}{kU} \quad (11)$$

Then, it is necessary to evaluate the validity of the constant frequency factor model. Therefore, we prepared a computer program based on constant frequency factor model with numerical analysis using MATHLAB. Figure 2 shows the predictions from above equations and compares them against measurements. It shows relatively good agreements. And the Simmons suggested a simple method to analyze the effects of stored energy on moderator temperature [1]. We could also design a code about it. From this, we can evaluate
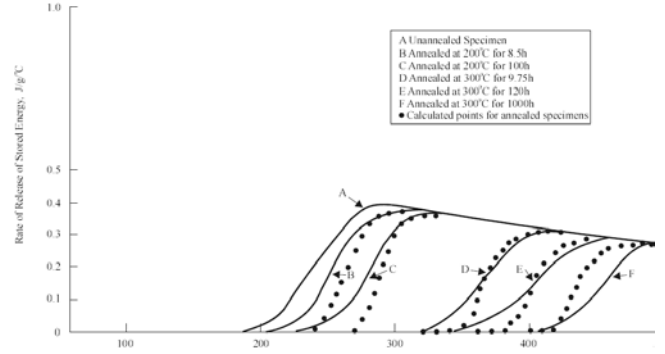

Figure 2 The effect of annealing at 200 and 300 for various times on the linear rise curve of graphite irradiated at 155 to a dose of 2.35×10^{20} n/cm^2.

the heat conduction and the distribution of temperature in graphite moderator. This is directly related to the safety assessment of reactor management.

3. Conclusions and Discussions

With increasing number of the reactors, there will be some necessitates for evaluating the release rate of stored energy.

We need to evaluate our code for using this to actual conditions. Therefore, we need to experiment with graphite in adjustable conditions and compare them with the theoretical values. For this, first we need investigation of Graphite material properties and selection. Next, the irradiation condition also should be decided.

From these, we are anticipating this can give a good estimation of the release rate of stored energy and heat conduction and temperature distribution in reactor component. And with these results, we can improve the theory.

Furthermore, we need to discuss the possible irradiation conditions for safety operation and how we can minimize the effects of stored energy release or how effectively we can remove the heat from reactor.

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