STORED ENERGY IN IRRADIATED SILICON CARBIDE — L. L. Snead and T. D. Burchell (Oak Ridge National Laboratory)

OBJECTIVE

A short discussion of the possibility for the release of stored energy (Wigner energy) from irradiated silicon carbide is presented. The objective of this work was to provide a basis to decide if further study into this area is warranted.

SUMMARY

This report presents a short review of the phenomenon of Wigner stored energy release from irradiated graphite and discusses it in relation to neutron irradiation silicon carbide. A single published work in the area of stored energy release in SiC is reviewed and the results are discussed. It appears from this previous work that because the combination of the comparatively high specific heat of SiC and distribution in activation energies for recombining defects, the stored energy release of SiC should only be a problem at temperatures lower than those considered for fusion devices. The conclusion of this preliminary review is that the stored energy release in SiC will not be sufficient to cause catastrophic heating in fusion reactor components, though further study would be desirable.

PROGRESS AND STATUS

The production of interstitial-vacancy pairs in graphite by neutron irradiation was postulated by E. P. Wigner in 1942. Later, it was pointed out by L. Szilard that when these defects recombine the defect formation energy of approximately 8 eV will be recovered (released). These two phenomenon became known as the Wigner Effect and the Szilard Complication, respectively. The potential danger of stored energy release during reactor operation was discovered early in the operation of the air cooled graphite production reactors at Windscale in the United Kingdom. To prevent the build-up of excessive amounts of stored energy the reactors were routinely thermally annealed to reduce the extent of the 200°C release peak. The Windscale reactors were successfully annealed on numerous occasions. However, the 1957 Windscale reactor 1 graphite fire was initiated by an anomalous stored energy release during a scheduled anneal.

Because of the wide scale use of graphite in gas cooled reactors a significant study was undertaken in the UK and the US into the stored energy release of irradiated graphite. It is now well understood that for low irradiation temperatures (<150°C,) the mobility for interstitial carbon is small and huge interstitial concentrations build-up between the graphite basal planes. For lower doses these defects remain as mono-interstitials though as the damage level is increased more complex groups are formed.[1] Because it is more difficult to recombine complex defects a temperature-dependent saturation occurs in the total stored energy available for release. If the temperature of the material is increased above the irradiation temperature the carbon interstitials increase in mobility and can recombine with interstitials releasing their formation energy. In recombining, approximately 8 eV is liberated leading to total stored heats of as much as 2,600 J/g (for graphite irradiated at 30°C.)[2] As the irradiation temperature is increased the equilibrium concentration of carbon interstitials falls off rapidly thereby reducing the total energy stored in the graphite. The decrease in the amount of stored energy accumulated is shown in Fig. 1.[3] While this figure does not show the saturation levels, it appears that there is less than 20% of the stored energy in material irradiated at 180°C as compared to materials irradiated at 25°C.

While a significant amount of energy can be stored in the crystal of irradiated graphite (and silicon carbide), the more critical issue is the kinetics of energy release. For the case of graphite irradiated at low temperature, and to low dose, the stored energy is released primarily in two peaks located at between 100 and 200°C. In order for graphite, or any material, to have an uncontrolled temperature increase due to the release of stored energy, the amount on energy release must exceed the specific heat at that temperature. As illustrated in Fig. 2,[3] the integral of the area between the energy release curve for an irradiated sample and the unirradiated specific heat can be used to calculate the stored-energy driven temperature rise. As

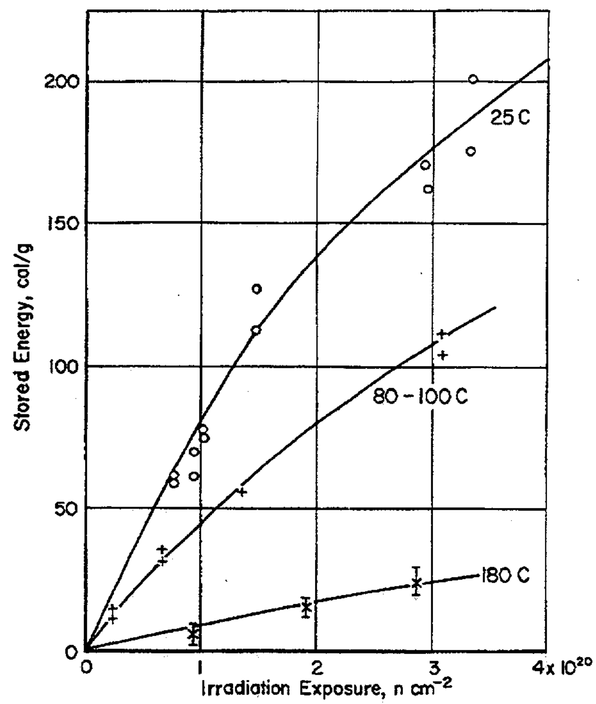

Figure 1. Stored energy in irradiated graphite.[3]

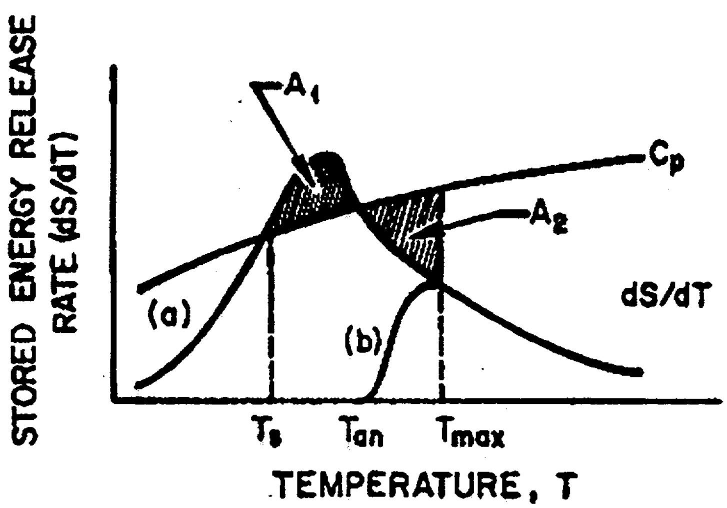

Figure 2. Schematic of stored-energy driven temperature rise.[3]

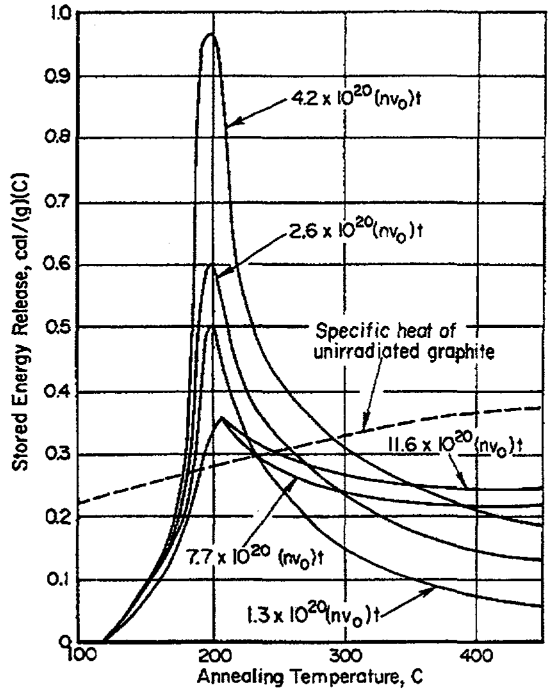

Figure 3. Stored energy release in graphite irradiated at 30°C.[4]

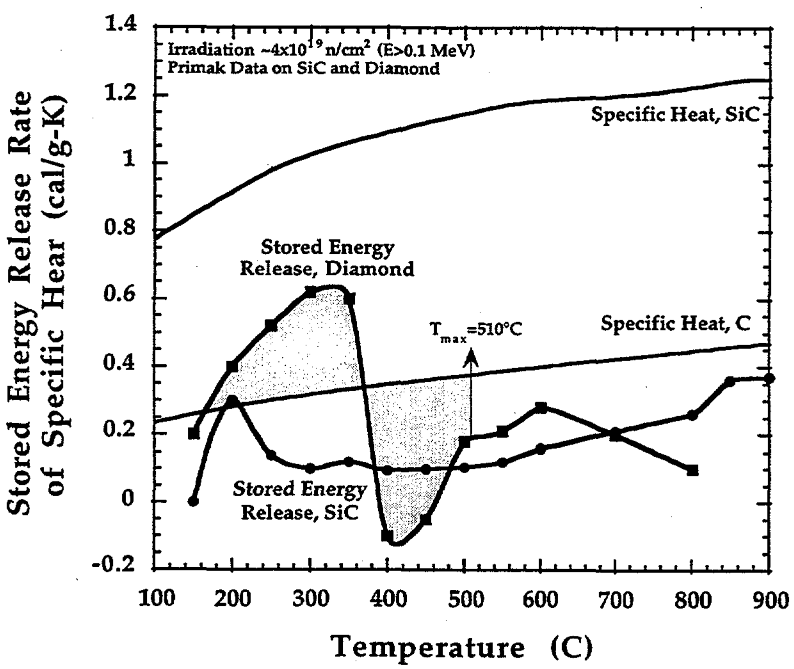

Figure 4. Energy release curves for diamond and silicon carbide.[5]

irradiation dose for graphite is increased and the defect structures become more complex, the release peaks broaden so that significant stored energy release occurs as high as 400-500°C. This is demonstrated in Fig. 3 for graphite irradiated in a fluence range of 1.3 and 11.6×10^{20} $(nv_o)t$.[4] It is seen in this figure that the potential sample temperature rise, as indicated by the integrated area above the specific heat, is greatest at the intermediate fluence of 4.2×10^{20} $(nv_o)t$. As the fluence increases from this point the energy release is substantially decreased due to the broadening of the release peaks.

The only published research on the release of stored energy in irradiated silicon carbide was carried out by Primak.[5] In this work, the energy release was measured for silicon carbide and diamond (which is very similar to graphite) using a differential thermal analysis apparatus at a heating rate of 20-40 degrees Celsius per minute. Single crystal SiC was irradiated in the cooled test hole facility of the Hanford reactor to a total dose of 255 MWd/at, or a "damaging" flux quoted as 4.3×10^{19} n/cm^2. The term "damaging" flux was described elsewhere[6] as 0.01 to ~10 MeV. Using a correction, and the 20% uncertainty in the fluence stated by Primak[5], the assumed E>0.1 MeV fluence for this study was $3.6 \pm 0.7 \times 10^{19}$ n/cm^2.

Plots the energy release data of Primak[5] and the specific heats of both carbon and silicon carbide are in Fig. 4. Two observations can be made from this data. First, the majority of the stored energy release of diamond occurs in the lower temperature regime as expected from previous work with graphite. In this case a stored energy driven temperature rise of approximately 125°C would occur for the diamond sample. Second, the data shows that the stored energy release for SiC at low temperatures is less than for diamond. It was also shown by Primak[5] that irradiated SiC has a much broader (and flatter) range in activation energies, which peaked above 3 eV as compared to the 1.5 eV peak of diamond.

The stored energy release characteristics of SiC will be a function of the total stored energy (dose and temperature dependent) and the release kinetics. For the stored energy rate (dS/dT) to exceed the specific heat of SiC requires both the accumulation of sufficient damage, and for its release over a relatively narrow temperature window. It is probably that Primak's data (~0.04 dpa) was for material below its saturation neutron damage dose. By applying the well established relationship between thermal conductivity and the total stored energy in graphites[7], and applying our experience with radiation damage in SiC, we are able to extrapolate Primak's data to a postulated saturation damage dose. If this is assumed to be the case for SiC, the saturation in stored energy should occur at, or less than, 0.1 dpa for temperatures under 200°C. The saturation of thermal conductivity by 0.1 dpa is demonstrated in Fig. 5 for irradiation temperatures in the range of 80-200°C.[8] To first order, the data of Primak can be scaled by a factor of two to estimate the amount of stored energy released in SiC at saturation. This is shown in Fig. 6. From the figure it is seen that a factor of two would yield an energy release well below the specific heat of SiC. A more pessimistic scenario is also shown in Fig. 6, where Primak's release curve is arbitrarily increased by a factor of five. Such a release would cause a temperature increase from about 240 to 300°C (T_{max}). It is important to note that this simple scaling does not account for any broadening in the release peaks which were discussed previously. However, this represents a conservative assumption as the effect would be to flatten the energy release curve.

CONCLUDING REMARKS

It appears that the release of Wigner energy is not a concern for using SiC as a structural material in fusion reactors for the following reasons.

(1) Existing data, while very limited, indicates that the energy release from SiC is less than that for graphite. Moreover, the significantly higher specific heat of silicon carbide compared to graphite suggests that the potential problem for SiC is considerably less than that of graphite. If Wigner energy release were a problem, an engineering solution such as annealing cycles (similar to those used to mitigate Wigner energy in gas cooled reactors) may be employed.

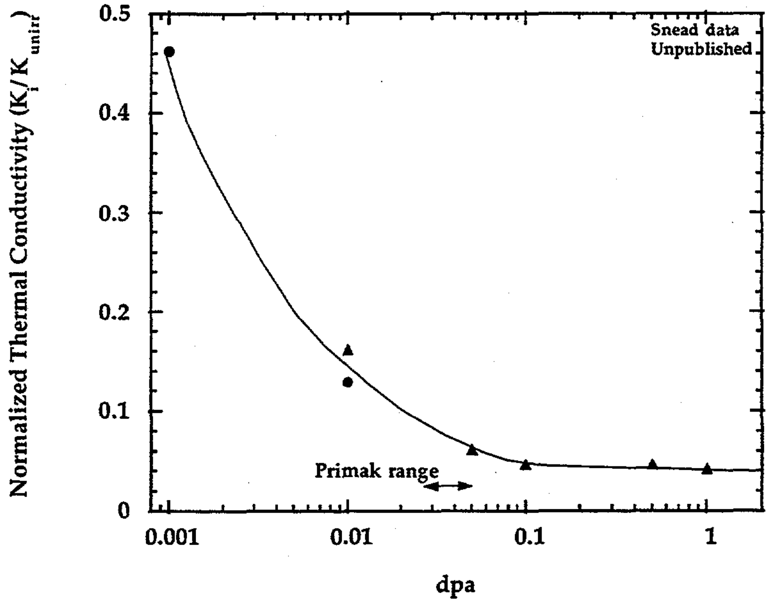

Figure 5. Thermal conductivity degradation for Morton CVD SiC under irradiation.[8]

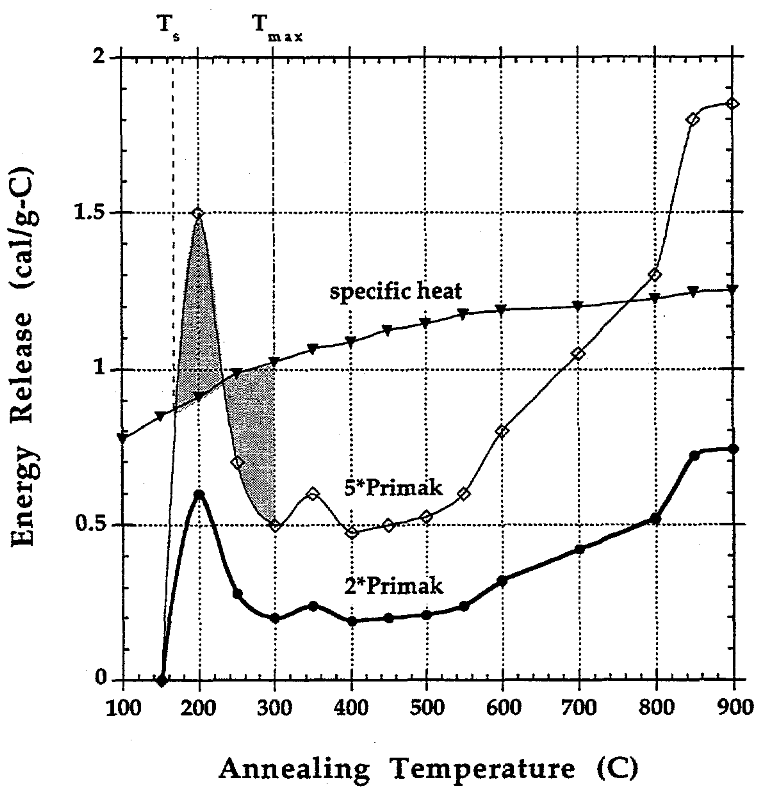

Figure 6. Assumed levels of stored energy release at saturation for SiC.

(2) The energy release peaks for SiC appear to be fairly broad and flat allowing the energy release due to recombination of defects to occur over a wide temperature range. This is in stark contrast to graphite, which releases the majority of its stored energy in the temperature range of 100 to 250°C. This former release characteristics suppresses spontaneous temperature rise in the case of SiC. Such behavior is probable for all ceramic materials because of the difference between the weak Van der Walls bonding of interstitials between basal planes of graphite and the covalent bonds in SiC or ionic bonding of other ceramics.

(3) It is unlikely that components of a fusion system which will see significant neutron flux will be at temperature less than 300°C. The data presented in this paper were in for materials irradiated somewhere between 80 and 150°C, and should therefore have significantly greater amounts of stored energy owing to the reduced interstitial mobility at these temperatures. For this reason, the total stored energy will be less at the elevated operating temperatures of fusion systems. However, if SiC is to be used for heating or diagnostic systems where it is cooled below 150°C, stored energy may be a consideration.

It is possible that the assumptions made for the saturation of stored energy of SiC based on Primak's data are in error, either because of to problems in using the analog between thermal conductivity saturation and stored energy saturation, or in determining the fluence of Primak's data. While it is unlikely that these errors will be large enough to alter our conclusion that stored energy in SiC is not a major issue in fusion systems, a series of simple experiments would confirm our conclusions.

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