PASSIVE SIC IRRADIATION TEMPERATURE MONITOR

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OBJECTIVE

The objective of this work is to examine the use of monolithic SiC as a passive irradiation temperature monitor using several methods.

SUMMARY

A new, improved passive irradiation temperature monitoring method was examined after an irradiation test at 627°C. The method is based on the analysis of the thermal diffusivity changes during post-irradiation annealing of polycrystalline SiC. Based on the results from this test, several advantages for using this new method rather than a method based on length or lattice parameter changes are given.

PROGRESS AND STATUS

Introduction

Over the past twenty years, several workers[1-5] have tested monolithic SiC bars for temperature monitoring in uninstrumented reactor experiments. Test methods are based upon the defect annealing behavior of neutron irradiated SiC; therefore they are out-of-reactor or passive determinations of the actual irradiation temperature. Typically after irradiation, small bars (one or two centimeters long) are subjected to an isochronal annealing schedule consisting of increasing temperature steps, each about one hour duration. When the sample is heated above the irradiation temperature, some of the irradiation defects recover and the remaining defects reach a new quasi-stable concentration dependent on the annealing temperature. Generally, changes in the macroscopic length or the lattice parameter are monitored between each temperature step as they recover toward their unirradiated values. Then a simple graphical approach which plots length (or lattice parameter) versus annealing temperature is used to obtain a fairly accurate prediction of the irradiation temperature. Nevertheless, about a 0.02% length change per anneal usually is observed, which for a bar of length 1.0 cm translates to a required precision in a length measurement of about ±0.1 μm - a very formidable task!

In irradiated SiC/SiC composite, 78% and 66% reductions in thermal diffusivity recently were reported for irradiations at 500° and 800°C, respectively.[6] After further heating to 1200°C, recoveries of almost 30% of the thermal diffusivity change then occurred. From these observations, it was apparent that a method for determining the irradiation temperature based on changes in the thermal diffusivity potentially would be more sensitive than when using length change methods.

Point defects are effective phonon scatterers, therefore irradiation defects should reduce the thermal diffusivity somewhat in proportion to their concentration. Following an annealing schedule similar to that used in the length change methods utilizing SiC, the thermal diffusivity should also recover toward its unirradiated values. Thus, a proposed method for predicting the irradiation temperature is patterned after the Isochronal Anneal Length Change (IALC) method and, hereafter, will be called the Isochronal Anneal Thermal Diffusivity (IATD) method.

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Initial results examinig the feasibility of using high purity, monolithic SiC and the IATD method for predicting the irradiation temperature is the subject of this report.

Experimental Procedure

Two small discs, either polycrystalline α-or ß-SiC, were received from Monbusho. Each disc was 1.0 cm diameter by about 2 mm thick with a bulk density greater than 97% of theoretical.

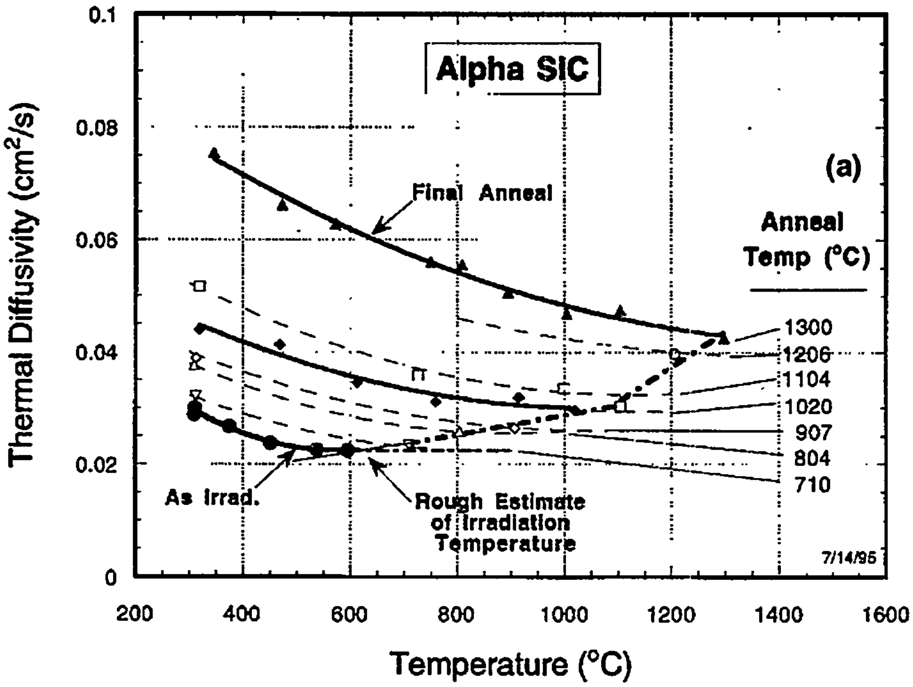
As part of the COBRA 1A1 experiment, the discs were irradiated in the EBR II breeder reactor in Idaho Falls over a period of 88.6 EFPD in subcapsule G05, which was a gas-gapped capsule designed to operate at 627°C. The fluence was 1.75×10^{22} n/cm^2 (E>0.1 MeV), equivalent to a dose of about 20 dpa-SiC. The samples were discharged from the reactor in April, 1993. The measured sample activity on contact was less than 3mR after a two year storage period.

The irradiated sample discs were mounted at the top of a low inertia, tungsten alignment tube that extended up into a tungsten mesh heating element inside a gas tight, water-cooled furnace wall. Then the samples were subjected to a series of one hour isochronal anneals in argon at temperatures increasing in about 100°C steps. The thermal diffusivity (TD), measured by the laser flash diffusivity technique and described elsewhere,[7] was measured periodically during the anneals and also at the end of each anneal after cooling the sample to a baseline temperature near 320°C. Importantly, the sample temperature could be rapidly changed in this system (at ≈20°C/min.) to the desired anneal temperature and then held constant to ±2°C during the anneals. To arrive at an average baseline TD value, at least five shots were made after the sample temperature had stabilized at 320°C. Otherwise, TD measurements were taken at about five to ten minute intervals during the one hour anneals to monitor the recovery kinetics. A curve was best-fit to this time dependent annealing data and the TD value determined at the end of each anneal was used as the anneal temperature TD value. The 320°C baseline temperature was chosen because the furnace and sample holder design geometry, made to accomodate radioactive samples, limited TD measurements to the 300° to 1500°C temperature range.

Results and Discussion

Figures 1(a-b) show the measured TD values as a function of temperature for the α- and ß-SiC samples, respectively. Initially, TD measurements were made on heating the irradiated samples up to about 600°C (data indicated as filled in circles); then the temperature was reduced back to the baseline temperature where the TD measurement was repeated. After this measurement cycle up to 600°C, which was somewhat below the irradiation temperature of 627°C, the baseline TD values remained unchanged, as expected for SiC with a quasi-stable as irradiated defect concentration. In both figures, open symbols indicate TD values determined at the end of the one hour anneal periods and following the anneals at the baseline temperature. After an intermediate and a final anneal at temperatures of ≈1000° and 1300°C, respectively, TD measurements were again made as a function of increasing temperatures to trace out the temperature dependence for that particular partially recovered state. These data are indicated with closed symbols and are connected with heavy solid lines in each figure. The TD temperature profiles for the other partially recovered states were estimated by tracing in lightly dashed curves roughly parallel to the solid curves. A darkened dot-dash line is shown traced through the anneal temperature TD values which represent the partially recovered states (anneal temperatures listed along the right-hand border in each figure). The intersection of this line with the extension of the "as irrad." curve, which showed no recovery, yields a rough estimate of the irradiation temperature of from 600° to 700°C. However, a more sensitive estimate can be made by comparing the changes in the TD at a low temperature reference where the effect of phonon scattering from point defects dominates the intrinsic phonon-phonon scattering. In our case, 320°C was selected for this reference.

In Figure 2, the 320°C TD values determined after each isochronal anneal are plotted versus the anneal temperature for both the α- and ß-SiC materials. The horizontal straight lines drawn through the initially measured 320°C TD values are characteristic of the fully irradiated defect state for each material. A third



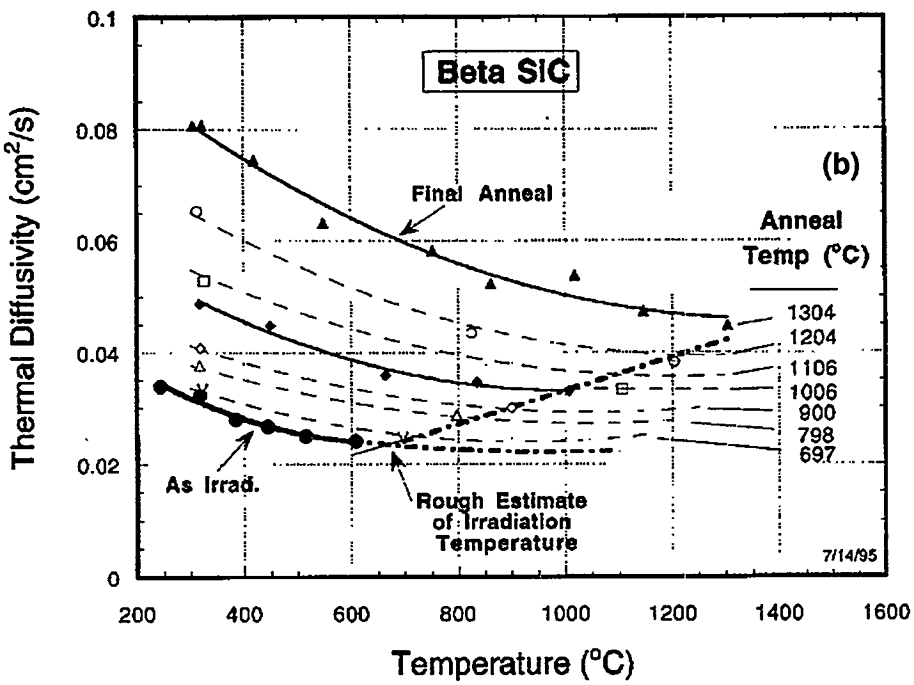


Figure 1(a-b) The thermal diffusivity temperature profiles for **(a)** α-SiC and **(b)** β-SiC after neutron irradiation to a dose of 20 dpa-SiC and after post-irradiation one hour isochronal anneals from 700° to 1300°C in ≈ 100°C steps.

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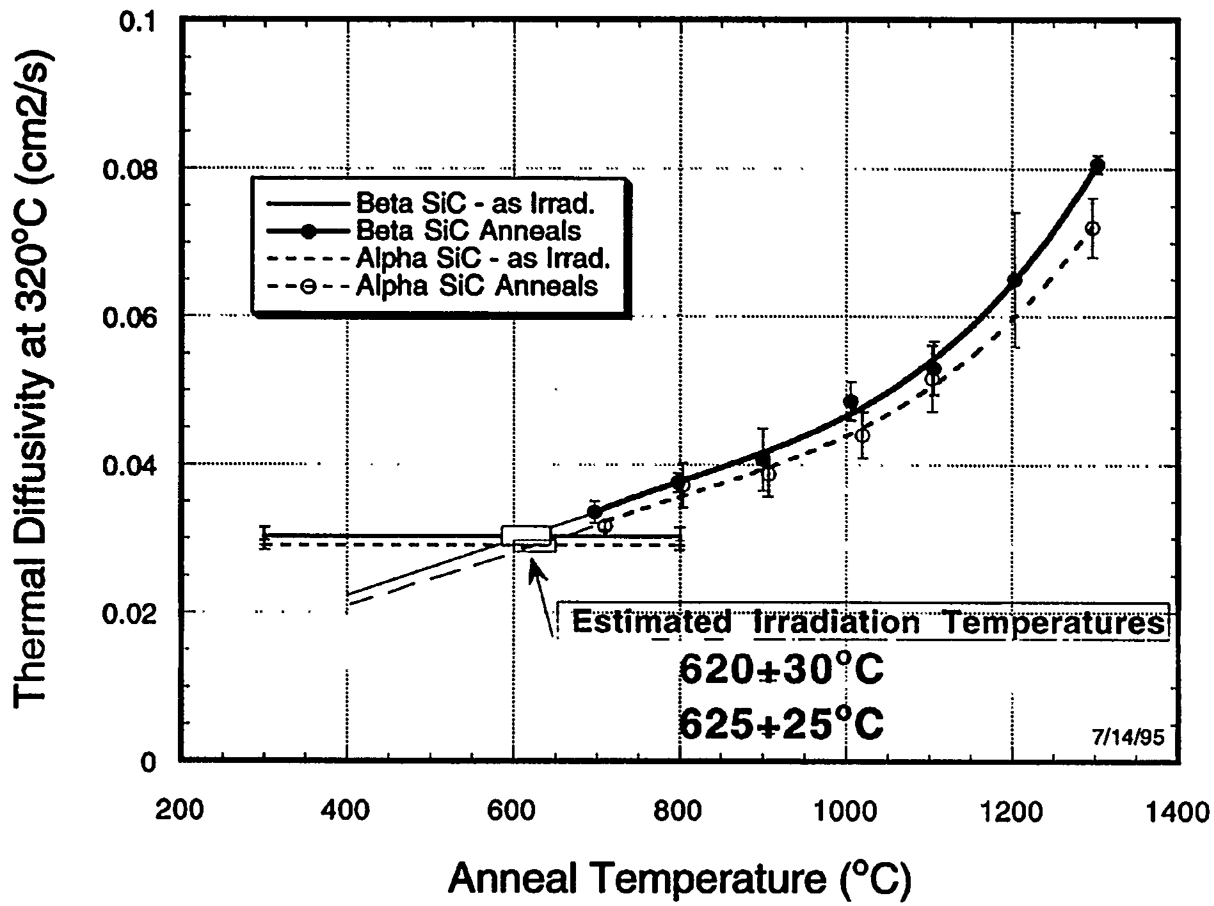

Figure 2 Estimate of the irradiation temperature using passive SiC temperature monitors and the isochronal anneal thermal diffusivity (IATD) method. For comparison of the thermal diffusivity changes due to partial recovery of the irradiation defect states after a series of isochronal anneals, the baseline temperature was taken as 320°C.

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order polynomial curve was best-fit to the 320°C TD values determined after each isochronal anneal. Above the irradiation temperature, these two curves are characteristic of the partially recovered defect states. The irradiation temperature is then estimated by the intersection of the straight line extension of the polynomial curve with its associated the horizontal baseline for the two materials. The two boxes reflect the uncertainties in the intersection points due to the uncertainties in the baselines as well as in the polynomial curve extensions. The irradiation temperatures estimated from the two intersection points are 625±25° and 620±30°C for the α- and ß-SiC materials, respectively. These predicted irradiation temperatures are in very close agreement with the design irradiation temperature of 627°C within subcapsule G05. Several other observations are worth noting.

First, the 300K TD values, estimated for each type SiC from the extensions of the "as irrad." curves from about 300° down to 27°C, were each about 0.045 cm^2/s. The phonon mean free path[7] estimated from this TD value is about 1 nm, which is a little over x2 lattice constants (l.c. = 0.436 nm for ß-SiC[8]). Therefore, the "as irrad." defect state (≈10^4 appm) is fairly close to saturation after irradiation to a dose of 20 dpa-SiC, even at 627°C.

Second, the magnitudes and shapes of the temperature profiles representing the partially recovered defect states were similar for each SiC phase. For such heavily irradiated SiC, the structural details appear to become dominated by the irradiation defect structures regardless of the original SiC phase structures. Thus, the curves representing the partially recovered or annealed states for the α- and ß-phases (in Figure 2) were almost identical.

Third, the selected isochronal annealing schedule (one hour anneals in 100°C steps) probably was not optimized for precisely determining the irradiation temperature. The TD measurements made as a function of time during the recovery anneals for temperatures up through 900°C indicated that the TD had not reached equilibrium values, but, in fact, was still increasing at the end of the one hour anneal. An annealing schedule designed to allow recovery to approach completion at each step should yield larger changes in the TD, thus a steeper slope in the curve representing the partially recovered states (less uncertainty in the intersection point). Perhaps using one or even two hour anneals with 50°C steps would allow a more accurate prediction of the irradiation temperature. Furthermore, even more sensitivity would be available if the baseline temperature were lower than 320°C, i.e., room temperature, for instance. Of course, neither the annealing kinetics, nor the temperature dependence of the TD values representing the partially recovered states need to be monitored once the optimum annealing schedule has been established.

Fourth, there appears to be a major difference between what is measured with the IALC method and what is measured with the IATD method. Even though vacancies and interstitials are generated in pairs during irradiation, they may have different influences on the observable physical properties. For instance, the associated shrinkage strain field of a vacancy must be smaller than the expansion strain field of an isolated interstitial since net swelling results from the accumulation of simple Frenkel pairs in SiC.[3] After neutron irradiation at intermediate temperatures (T≤900°C), the residual interstitial type defects primarily consist of a relatively high concentration of small interstitial clusters (≈1-5 nm).[9] For irradiations at higher temperatures or to higher fluences (≥10^{22} n/cm^2), the interstitial defects start to interact and form into more orderly fault or dislocation type loops as required to preserve the covalent Si-C tetrahedral bonding.[10] The interstitial loops have a lower strain energy per defect and therefore contribute little to the macroscopic swelling of SiC. This interpretation is still controversial and other explanations exist, i.e., that of Huang and Ghoneim.[11] Also, the demarcation temperature of 900°C is approximate, as it certainly depends on the irradiation conditions, particularly the neutron fluence. Meanwhile, the mobility of vacancies is much smaller than that of interstitials, so they remain relatively stable and isolated. Thus, the IALC method primarily monitors the decreasing size or concentration of these smaller, cluster type interstitial defects as these defects anneal either by combining into larger loop type interstitial defects (resulting in a smaller expansion strain field per interstitial) or completely disappear by recombination with the residual vacancies. Since the formation of small, cluster type interstitial defects predominates the formation of the larger, loop type interstitial defects only for irradiations below about 900°C, the IALC method is limited in application to this temperature range.

It is likely that the strain fields of relatively isolated point defects, such as impurity atoms or vacancies, are more effective phonon scatterers than are the more extensive strain fields of the clustered interstitial defects, and certainly more effective than the quite extensive interstitial loop structures. Thus, the IATD method primarily monitors the decreasing concentration of vacancies which only anneal out by recombination with mobile interstitials below about 1300°C.[12] Above 1300°C, vacancy mobility becomes sufficient for the vacancies to anneal out to grain boundaries or other sinks or to combine with other vacancies. In the latter case, vacancy clustering can lead to void or bubble formation, especially if a bubble stabilizing gas such as helium is available within the lattice.[12] Since the TD is primarily sensitive to the vacancy concentration, it appears probable that the IATD method can be used to predict the irradiation temperature to a temperature higher than 900°C. This would be extremely useful for currently planned materials tests of the irradiation performance of SiC or SiC/SiC in the 800° to 1000°C temperature range.

Another significant advantage of an IATD method over IALC methods would be, once the irradiated SiC sample is mounted, all thermal diffusivity measurements and anneals would take place in situ, thus no additional handling of a radioactive sample would be required between anneals. Finally, the use of a high-purity SiC sample, such as one made from Morton CVD/SiC, should further improve the sensitivity of the IATD method since recovery will only involve the induced irradiation defects.

The main disadvantage of either the IATD or the IALC methods is that both methods are sensitive to the temperature during the final period of irradiation due to saturation effects at doses of only a few tenths of a dpa, which may be equivalent to only a few days irradiation in a fast reactor.

CONCLUSIONS

As part of the U.S./Japan collaborative program to study the effects of neutron irradiation on fusion reactor candidate materials (NIMS/ Monbusho), a new improved method for determining the irradiation temperature of an uninstrumented experiment was examined. The new method is called the Isochronal Anneal Thermal Diffusivity (IATD) method. In the IATD method, recovery in irradiated SiC is monitored by measuring the change in the thermal diffusivity (TD) after a series of postirradiation isochronal anneals. The TD is a more sensitive measure of the recovery than length change, so the IATD method is a more sensitive predictor of the irradiation temperature than the Isochronal Anneal Length Change (IALC) method. Other advantages for using the IATD method over the IALC method are its potential for monitoring the annealing kinetics and the reduced handling of a radioactive sample. Finally, the IATD method may be usable to predict irradiation temperatures as high as 1200°C, whereas IALC methods appear to be limited to predicting irradiation temperatures less than about 800°C.

FUTURE WORK

1. Two 20 mm bars made from the same materials as the discs used in the IATD method study were irradiated under identical conditions (≈20 dpa at 627°C) in COBRA 1A1. The bars will be analyzed using the IALC method, and the results will be compared to the IATD results. Also, the defect structures in the SiC's for the as irradiated and for the partially recovered conditions will be analyzed by TEM.

2. Two bars and one disc of very high purity SiC and of theoretical density (Morton CVD/ß-SiC) have been irradiated at about 800°C in the COBRA 1A2 experiment to an extremely high dose of ≈80 dpa-SiC. These samples were discharged from the EBR II reactor after 337 EFPD. Their annealing characteristics as well as their utility as an irradiation temperature monitor (using both the IALC and IATD methods) for such extreme conditions (high dose and temperature) will be examined.

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