DILATOMETRY OF IRRADIATED SILICON CARBIDE AS A METHOD FOR DETERMINATION OF IRRADIATION TEMPERATURE —Y. Katoh, A. Clark, K. Ozawa (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this work is to evaluate applicability of temperature dependent dimensional evolution of cubic silicon carbide for determination of irradiation temperature.

SUMMARY

Dilatometry-based thermometry using SiC appeared to be consistent with the result of conventional electrical resistivity-based technique within the irradiation conditions examined in this work, ~673K (~400 ºC) to ~1073K (~800ºC). The maximum difference in irradiation temperatures determined by these two methods was ~20K. This method appeared to be as reliable as the conventional electrical resistivity-based technique within the irradiation conditions examined in this work.

PROGRESS AND STATUS

Introduction

Thermal expansion of irradiated SiC will be affected by defect annealing when heated beyond the temperature at which the sample was irradiated. Linear thermal expansion measurement may be used for passive thermometry of reactor irradiation, if the onset of such annealing can be explicitly determined.

Benefit of dilatometry-based thermometry will be the fact that the irradiation temperature may be determined in one measurement run which typically can be completed within a day or over-night. The conventional thermometry method involving repeated isochronal annealing processes and electrical resistivity measurement at a fixed temperature typically takes weeks to complete.

Moreover, there have been only very limited reports on the effect of irradiation on the thermal expansion behavior of SiC. The result obtained here may be scientifically and technologically valuable.

Experiment

The material used was high purity (manufacturer-claimed purity >99.999%, nitrogen as the primary impurity) chemically vapor-deposited (CVD) SiC manufactured by Rohm & Haas Advanced Materials (now Dow Chemical Co.). The irradiated samples used in this study were a part of fixture, called “retainer,” used in the bend stress relaxation irradiation creep experiment performed in the fusion US-Japan TITAN program. The nominal irradiation temperatures and doses are shown in Table I.

Table I: Summary of nominal irradiation conditions, swelling, and the dilatometric annealing onset temperatures.

Capsule ID/ Specimen	Tirr (nominal)	Dose (dpa)	Linear Swelling	Ramp Rate (K/min.)	Defect Ann. Temp. per Dilatometry	Defect Ann. Temp. per ER*
M02	300ºC	0.11	0.50%	2.5	~400ºC	~390ºC
				0.25	~380ºC	
M05	500ºC	0.94	0.46%	2.5	~540ºC	~540ºC
M07	800ºC	1.38	0.25%	2.5	~760ºC	~740ºC

*ER: Electrical resistivity

The original length of the retainers was 40 mm. Linear swelling values in the above table were determined by measuring the change in length.

The dilatometry was performed up to about 1250K at a constant ramp rate of 2.5 K/min. for one sample from each capsule. Then a replicate sample from M02 (M02-2 hereafter) was measured to about 750K at the ramp rate of 0.25 K/min.

The other CVD SiC piece from the fixture, called “hub,” was subjected to the routine thermometry analysis. The pair of retainer and hub were irradiated side-by-side both pressed to the internal surface of the same molybdenum sleeve during irradiation. Therefore their irradiation temperatures are expected to be exactly same. The routine thermometry analysis is based on measurement of electrical resistivity at room temperature following isochronal annealing. The annealing time at each temperature step was 30 minutes.

Results and Discussion

All samples exhibited clear deflections in the length-temperature plots, Figs. 1 (a) through (c), approximately corresponding to the nominal irradiation temperatures. Specifically, the onset temperatures of apparent defect annealing during heating were 673, 813, and 1033K (400, 540, and 760ºC) as compared to the nominal irradiation temperatures of 573, 773, and 1073K (300, 500, and 800ºC) for M02, M05, and M07, respectively. The results are summarized in Table I.

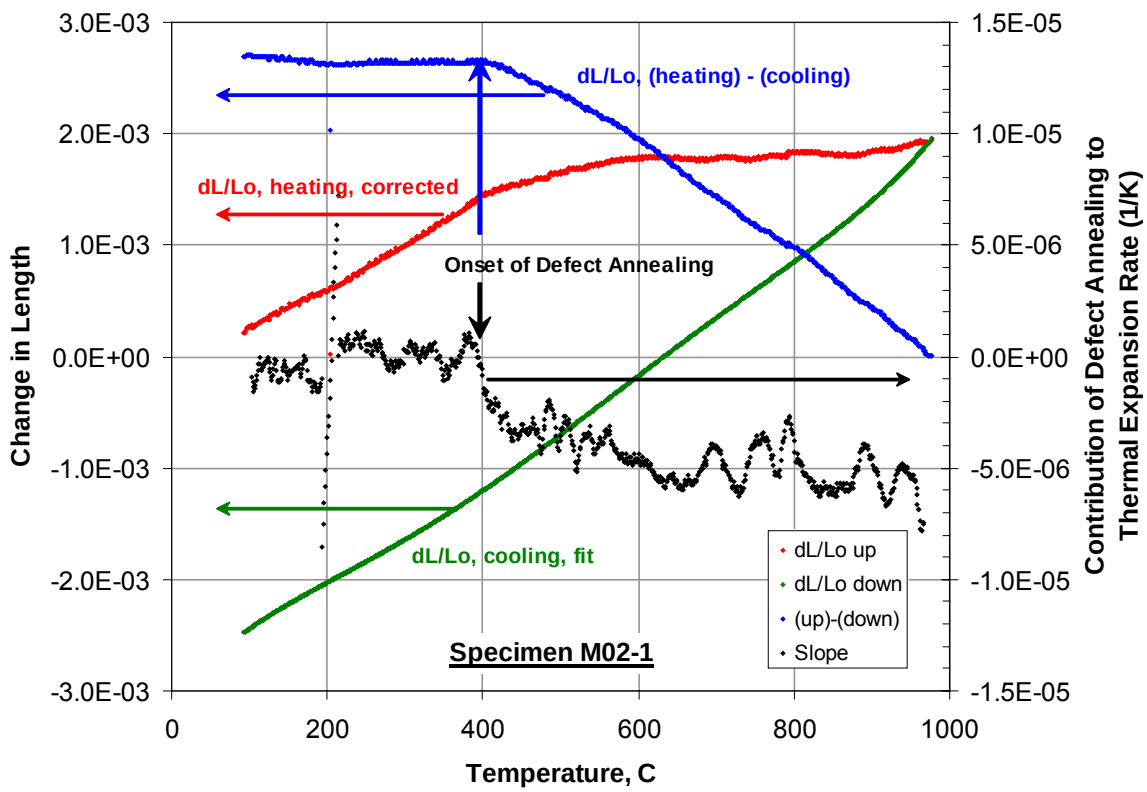

Figure 1: (a) Fractional change in length of M02 CVD SiC sample as a function of temperature at a constant ramp rate of 2.5 K/min. Red: initial heating; Green: cooling after initial heating; Blue: difference between initial heating and cooling; Black: derivative of the differential length change.

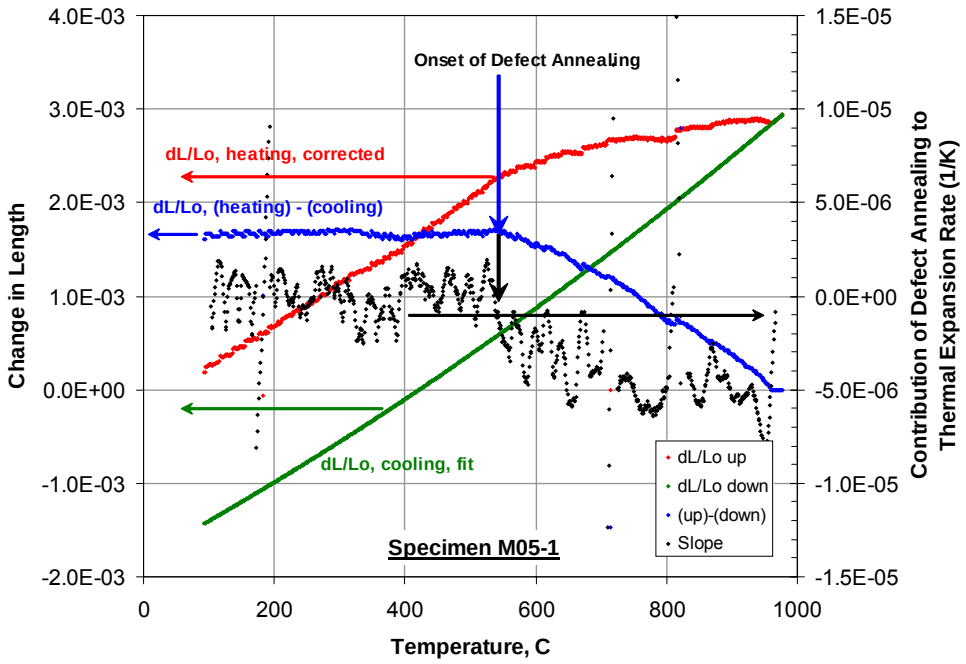

Figure 1: (b) Fractional change in length of M05 CVD SiC sample as a function of temperature at a constant ramp rate of 2.5 K/min. Red: initial heating; Green: cooling after initial heating; Blue: difference between initial heating and cooling; Black: derivative of the differential length change.

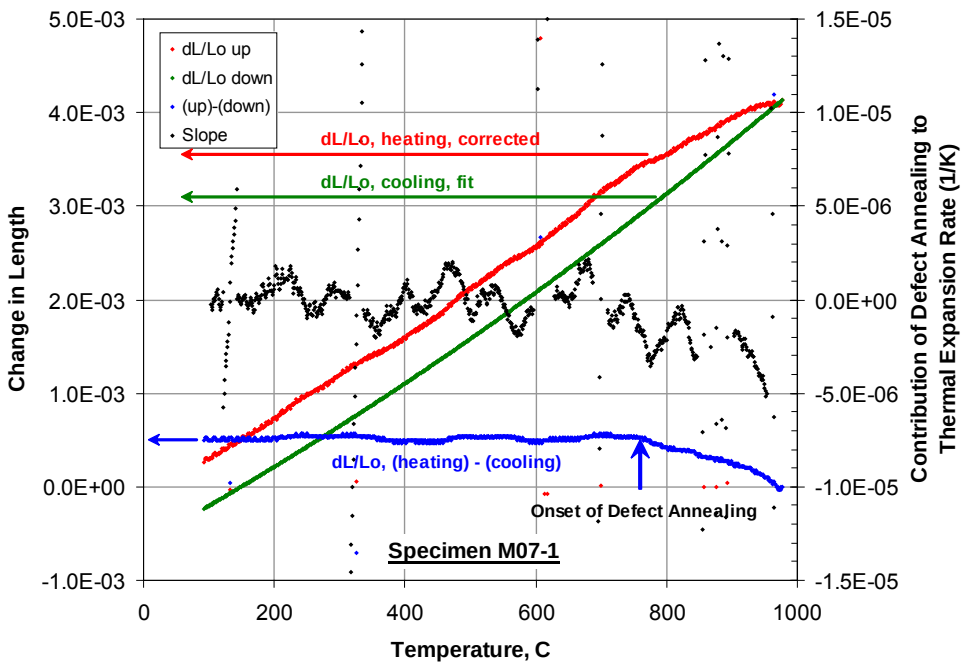

Figure 1: (c) Fractional change in length of M07 CVD SiC sample as a function of temperature at a constant ramp rate of 2.5 K/min. Red: initial heating; Green: cooling after initial heating; Blue: difference between initial heating and cooling; Black: derivative of the differential length change.

Figure 2 shows the result of dilatometry run at the slower ramp rate, 0.25 K/min., using the M02-2 sample. The onset of defect annealing in this case appeared slightly less clear due to the noise, but it is certainly in the vicinity of 653K (380ºC). This indicates about -20K difference from the annealing onset temperature at a ramp rate 2.5 K/min.

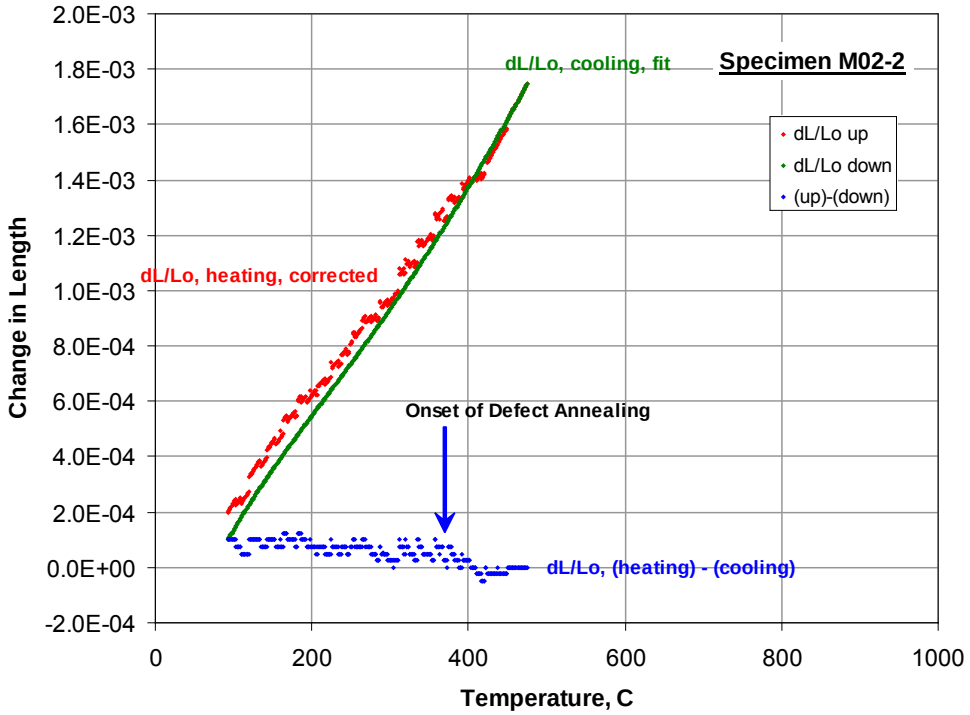

Figure 2: Fractional change in length of M02-2 CVD SiC sample as a function of temperature at a constant ramp rate of 0.25 K/min. Red: initial heating; Green: cooling after initial heating; Blue: difference between initial heating and cooling.

Figure 3 shows the electrical resistivity at room temperature as a function of annealing temperature. The onset temperature of defect annealing in this process appeared to be ~390ºC for M02, which falls between the defect annealing onset at ramp rates 2.5 K/min. and 0.25 K/min. during dilatometry. The onset temperatures of defect annealing for M05 and M07 were determined to be ~813K (~540ºC) and ~1013K (~740ºC), respectively, by the electrical resistivity at room temperature. These are also within ±20K of the onset defect annealing determined by dilatometry, Fig. 4.

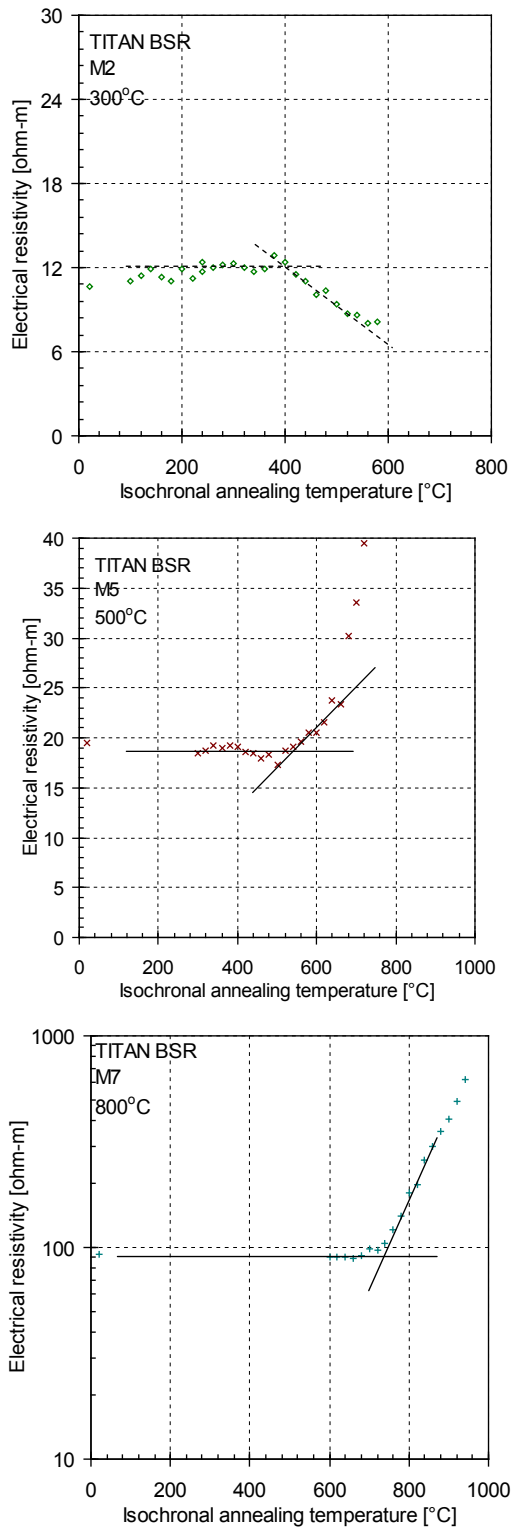

Figure 3: Isochronal annealing of room temperature electrical resistivity of CVD SiC samples, top: M02, showing defect annealing onset at ~663K (~390ºC); middle: M05,~813K (~540ºC); bottom: M07, ~1013K (~740ºC).

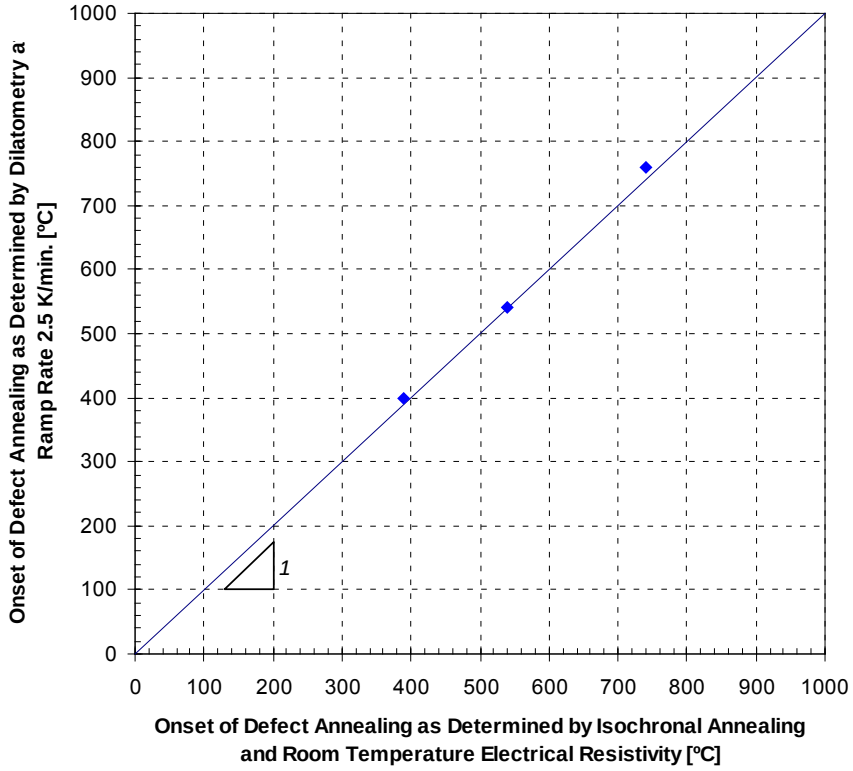

Figure 4: Correlation between the irradiation temperatures determined by dilatometry and isochronal annealing with intermittent room temperature electrical resistivity measurement.

The time required for thermal equilibration of the entire specimen housing is typically <5 minutes for the dilatometer used, thus the potential maximum shift in annealing temperature at the ramp rate of 2.5 K/min. will be 12.5K as compared to the pseudo-static annealing. The influence of ramp rate on defect annealing behavior is not known but may be contributing as well. In the routine thermometry practice, the isochronal annealing time is 30 minutes. In the present dilatometry runs, the temperature changes during 30 minutes are 75K at 2.5 K/min. and 7.5K at 0.25 K/min. However, assuming as an example that the responsible annealing process has an activation energy of 1.5 eV, temperature increase by 50K in the temperature range 573-673K will increase the annealing rate approximately by an order. Thus the difference in ramp rate between 2.5 and 0.25 K/min. may impose a significant shift in the apparent temperature dependent annealing behavior, although it is unlikely that the shift reaches many tens of Kelvin in most cases. On the other hand, typical error in determining the annealing onset temperatures from charts (Figs. 2 and 3) is anticipated to be within 10K. Therefore, it is reasonable to assume that the observed ~20K difference in the annealing onset temperature between the two ramp rates is contributed by all of the equipment thermal equilibration time, the rate of defect annealing in relation with the equilibration time, and the error in reading the onset temperature.

Anticipated error range in determining irradiation temperature based on the conventional electrical resistivity-based passive thermometry is typically ±20K of the actual irradiation temperature.[1] To achieve similar accuracy with this dilatometry-based thermometry technique, ramp rates smaller than ~2.5 K/min. are recommended. Limited yet significant improvement in accuracy may be achieved by further decreasing the ramp rate.

Reference

[1] L.L. Snead, A.M. Williams, A.L. Qualls, Revisiting the Use of SiC as a Post Irradiation Temperature Monitor, in: M.L. Grossbeck, T.D. Allen, R.G. Lot, A.S. Kumar (Eds.) The Effects of Radiation on Materials: 21st International Symposium, ASTM STP 1447, ASTM International, West Conshohocken, PA, 2003.