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Method for analyzing passive silicon carbide thermometry with a continuous dilatometer to determine irradiation temperature ☆

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ABSTRACT

Silicon carbide is used as a passive post-irradiation temperature monitor because the irradiation defects will anneal out above the irradiation temperature. The irradiation temperature is determined by measuring a property change after isochronal annealing, i.e., lattice spacing, dimensions, electrical resistivity, thermal diffusivity, or bulk density. However, such methods are time-consuming since the steps involved must be performed in a serial manner. This work presents the use of thermal expansion from continuous dilatometry to calculate the SiC irradiation temperature, which is an automated process requiring minimal setup time. Analysis software was written that performs the calculations to obtain the irradiation temperature and removes possible user-introduced error while standardizing the analysis. This method has been compared to an electrical resistivity and isochronal annealing investigation, and the results revealed agreement of the calculated temperatures. These results show that dilatometry is a reliable and less time-intensive process for determining irradiation temperature from passive SiC thermometry.
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1. Motivation

The effect of irradiation on materials properties is a critical field of study for materials usage in both fission and fusion systems for energy production. Understanding the underlying mechanisms requires knowledge of the irradiation conditions, i.e., temperature, flux, neutron energy spectrum, etc. In many test reactors the neutron flux and energy spectrums are well understood, but the irradiation temperature can be more difficult to determine.

Materials irradiations in the flux trap region of the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR) allow for a high neutron flux and wide range of irradiation temperatures. The materials are located in specially designed capsules that have an internal diameter of 4.5–6 mm and an internal length of 48–50 mm, and eight capsules can be vertically stacked in the active length of the core. In a limited number of experiments, a single capsule has been designed that runs the entire length of the active core. Each capsule is individually designed in the ANSYS heat transfer analysis program, and dimensions of the specimen holder, gas gap, and fill gas are modified to obtain the desired irradiation temperatures. These designs still require either an active or passive method to determine whether the design temperature was achieved, or what temperature was achieved. The preferred method of temperature measurement is the use of thermocouples that are continuously monitored (active) so modifications to the fill gas can be made to achieve and maintain the design temperature. But, the small internal volume and the high cost for active instrumentation can be deterrents to using online temperature measurements. Therefore many times the use of a passive temperature monitor (TM) is utilized, specifically chemical vapor deposition (CVD) silicon carbide (SiC). The main disadvantage of the passive TMs is that final analysis only provides an indication of the

Abbreviations: SiC, silicon carbide; XRD, X-ray diffraction; TM, temperature monitor; DMTR, Dounreay Materials Testing Reactor; DFR, Dounreay Fast Reactor; JRR-2, Japan Research Reactor No.2; JMTR, Japan Materials Testing Reactor; ORNL, Oak Ridge National Laboratory; HFIR, High Flux Isotope Reactor; LAMDA, Low Activation Materials Development and Analysis Laboratory; CTE, coefficient of thermal expansion; CVD, chemical vapor deposition.

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irradiation temperature near the end of irradiation. There are also limits on the temperature/fluence range that SiC can be used, since either amorphization or void swelling would make temperature analysis impossible. These passive monitors may not be suitable for testing environments where the gamma heating may be continuously changing, but in HFIR the heat generation rate remains nearly constant so a time-averaged temperature is acceptable.

The historical methods of determining temperature from the SiC TMs utilized isochronal annealing for 0.5–2 h at each temperature step, where it is recommended that annealing steps be in increments of <20 °C for values within ±100 °C of the designed irradiation temperature. Larger steps are used outside this temperature range. This type of analysis is very time and labor intensive. In most materials irradiation programs at ORNL there are many tens of TMs that must be analyzed, depending on program size and number of capsules, and most HFIR programs have capsules at different design temperatures. Hence, these requirements provide the motivation to develop a methodology to determine irradiation temperature from TMs using a dilatometer.

2. Background

Passive thermometry has been investigated since the early 1960's, when Pravdyuk et al. [1] first observed that radiation damage in 6H SiC would start to thermally anneal out once the annealing temperature surpassed the irradiation temperature. They observed the recovery of radiation damage from the change in the crystal lattice volume, measured with X-ray diffraction (XRD), where there was little change in the crystal volume for annealing below irradiation temperature but a dramatic decrease in volume when the annealing temperature surpassed the irradiation temperature. Inspired by these results, Pravdyuk et al. [1] proposed using SiC as passive measurement of irradiation temperature.

Some of the first work to investigate and provide better quantification of SiC as a passive TM was conducted at Dounreay, in the Materials Testing Reactor (DMTR) [2] and the fast reactor (DFR) [3]. The effect of irradiation on the properties of porous β-SiC (2.22 and 2.12 g/cm^3 where 3.21 g/cm^3 is fully dense) was also first investigated at DMTR. The irradiations were performed at 250 °C, 475 °C, and 700 °C, included three thermocouples, and used electrical heating to maintain the thermocouple temperature. Researchers at DMTR [2] were the first to report that the radiation-induced swelling and the irradiation-induced decrease of thermal conductivity both saturate at a low dose without any phase change. They also demonstrated that isochronal annealing achieved the maximum reduction of irradiation-produced defects after 30 min of annealing time and that the final damage is characteristic of the end-of-irradiation temperature.

Researchers at DFR [3] investigating the effect of irradiation on graphite were the first to include SiC as a TM. The changes in length and thermal conductivity were measured after 1 h of isochronal annealing in a vacuum. The DFR results agreed with the DMTR results, but more importantly, the effect of having a lower irradiation temperature at the end of the irradiation was also observed. One DFR shutdown took approximately 11 h, and during the post-irradiation annealing, two kinks were observed in the length change, one at the irradiation temperature (470 °C) and one around 250–300 °C, approximately the average temperature during the shutdown process. The formation of two kinks will only occur when the end-of-irradiation temperature is lower than the main temperature. A higher end-of-irradiation temperature will anneal out the main damage and result in the TM showing annealing behavior related to the higher end-of-irradiation temperature.

More irradiations were performed at DFR [4], and with this increase in data, a near one-to-one agreement was found between the temperature from SiC annealing and the temperature measured with thermocouples. When measuring the TM length, errors of ±15–30 °C for temperatures below 750 °C and ±100 °C for temperatures above 1000 °C were found. Temperature confidence was increased by measuring the lattice parameter, which decreased the error to ±15 °C at 750 °C and ±25 °C at 1000 °C. The researchers also investigated the use of continuous dilatometer measurements as a way to reduce the labor-intensive nature of the isochronal annealing, but they noted that it was difficult to determine the temperature at which annealing began. The difficulty with the dilatometry most likely stemmed from a lack of available high-resolution equipment at the time.

Suzuki et al. [5] investigated different techniques that could be used to determine the irradiation temperature from TMs. They irradiated both powder and sintered rods of β-SiC in Japan Research Reactor No.2 (JRR-2) and in the Japan Materials Testing Reactor (JMTR). In the JRR-2 experiments, the SiC was located between two thermocouples, while in the JMTR, a sodium–potassium coolant separated the specimen and thermocouples. The four techniques they investigated included isochronal annealing with XRD lattice measurements and electrical resistivity, *in-situ* high-temperature XRD, and dilatometry. Their results for the 500 °C irradiation in JRR-2, with the thermocouples at the ends of the SiC, found that the isochronal annealing and XRD resulted in a temperature of 490 °C ± 20 °C. The *in situ* XRD annealing estimated a temperature of 510 °C ± 20 °C, and the isochronal annealing and resistivity obtained a temperature of 480 °C ± 20 °C. The dilatometry overestimated the temperature by 20 °C–80 °C, but they noted that the high heating rate of 5 K/min could have caused some of the overestimate.

Palentine [6] was the first to apply linear regression to the annealing data to determine the irradiation temperature. He applied this analysis to all the temperature data produced by the DMTR and DFR irradiations [2–4]. This analysis determined that agreement between the thermocouple and the TM temperature was not one to one and that the TM temperature overestimated the irradiation temperature. Unfortunately, in all these irradiations the SiC was not in direct contact with the thermocouples, and it is unclear where the thermocouples were in relation the SiC. Hence, it is possible that the temperature experienced by the SiC was different from the thermocouple-measured temperature. Without detailed heat-transfer models, questions regarding this nonlinear agreement cannot be answered.

More recently, Snead et al. [7] proposed that the disagreement between thermocouple temperatures and TM temperatures reported by Palentine was most likely due to errors inherent to measurement accuracy. Additionally, it was discussed that the SiC used in the previous work was hot pressed or low density, and recent work has indicated that residual grain boundary elements can cause differential swelling. They showed that density measurements (using a density gradient column), thermal diffusivity, and electrical resistivity measurements, in conjunction with isochronal annealing, are all suitable techniques for TM analysis.

3. Experimental

The TMs in use at ORNL are high-density chemical vapor deposition (CVD) β-phase SiC (cubic structure) from Rohm and Haas® (now Dow Chemical Company®). CVD-SiC is desirable for TMs because of the high density and the reduction of the residual grain boundary elements, which can cause differential swelling, as discussed in [7]. The size of the TMs is adjusted according to the available internal dimensions of each capsule, but a typical TM is 45 mm × 3 mm × 1 mm. The TMs are analyzed in the Low Activation Materials Development and Analysis Laboratory (LAMDA) with a NETZSCH DIL 402CD horizontal dual pushrod dilatometer. Before each run the vacuum-tight dilatometer is pumped down, then back