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# Use of Silicon Carbide Monitors in ATR Irradiation Testing

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# USE OF SILICON CARBIDE MONITORS IN ATR IRRADIATION TESTING

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## ABSTRACT

In April 2007, the Department of Energy (DOE) designated the Advanced Test Reactor (ATR) a National Scientific User Facility (NSUF) to advance US leadership in nuclear science and technology. By attracting new users from universities, laboratories, and industry, the ATR will support basic and applied nuclear research and development and help address the nation's energy security needs. In support of this new program, the Idaho National Laboratory (INL) has developed in-house capabilities to develop, fabricate, test, and qualify new and enhanced temperature sensors for irradiation testing. Although most efforts emphasize sensors capable of providing real-time data, selected tasks have been completed to enhance sensors for irradiation locations where instrumentation leads cannot be included, such as drop-in capsule and Hydraulic Shuttle Irradiation System (HSIS) or 'rabbit' locations. For example, silicon carbide (SiC) monitors are now available to detect peak irradiation temperatures between 200°C and 800°C in reactor locations where instrumentation leads cannot be used. Based on an electrical resistance measurement approach, specialized equipment installed at INL's High Temperature Test Laboratory (HTTL) and specialized procedures were developed to ensure that accurate peak irradiation temperature measurements can be inferred from SiC monitors irradiated at the ATR. Comparison examinations were completed by INL to demonstrate this capability, and several programs currently rely on SiC monitors for peak temperature detection. This paper discusses the use of SiC monitors at the ATR, the process used to evaluate them at the HTTL, and presents representative temperature data inferred from SiC monitors.

*Key Words*: silicon carbide (SiC) monitor, high temperature irradiation testing

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# 1 INTRODUCTION

Although thermocouples are used to provide real-time temperature indication in instrumented lead tests performed at materials and test reactors (MTRs), melt wires or paint spots are often included in such tests as an independent technique of detecting peak temperatures incurred during irradiation. In addition, less expensive static capsule and Hydraulic Shuttle Irradiation System (HSIS) or “rabbit” tests, which have no leads attached for real-time data transmission, often rely on melt wires and paint spots as a post-irradiation technique for peak temperature indication. Melt wires and paint spots are limited in that they can only detect whether a single temperature is or is not exceeded. However, silicon carbide (SiC) monitors, which can also be used to detect peak irradiation temperatures, are advantageous because a single monitor can be used to determine the peak temperature that may be reached within a relatively broad range. As part of the process initiated through the Advanced Test Reactor National Scientific User Facility (ATR NSUF) program to make SiC temperature monitors available for experiments, a capability was developed at the Idaho National Laboratory (INL) to complete post-irradiation evaluations of these monitors.

SiC monitors may be used to detect peak temperatures that occur during irradiation through evaluation of changes in electrical resistivity, thermal diffusivity, density, thermal conductivity, or through the use of X-ray line broadening to detect lattice parameter changes. INL selected the electrical resistance measurement approach for detecting peak irradiation temperature based on recent efforts by Oak Ridge National Laboratory (ORNL)[1][2] indicating that this approach provides more accurate measurements. This INL approach was used in the post-irradiation evaluation of SiC temperature monitors recently irradiated in the ATR and evaluated at the INL High Temperature Test Laboratory (HTTL) [3] as part of a University of Wisconsin Pilot Project [4].

This paper discusses the use of SiC monitors at the ATR, the process used to evaluate them at the INL HTTL, and presents measurements taken using SiC monitors as part of the University of Wisconsin Pilot Project. Section 2 gives historical information in the use and development of SiC monitors. Section 3 provides overview information about the ATR. Section 4 discusses the equipment and processes used at the HTTL to evaluate the SiC monitors and Section 5 presents results from this evaluation.

# 2 BACKGROUND

The use of electrical resistivity measurement techniques to infer peak irradiation temperatures from SiC monitors is relatively new. There are no American Society for Testing and Materials (ASTM) standards governing the evaluation of SiC temperature monitors using electrical resistivity techniques.

Since the early 1960s, SiC has been used as a post-irradiation temperature monitor. Pravdyuk[5] first reported that neutron induced lattice expansion of SiC annealed out when the post-irradiation annealing temperature exceeds the irradiation temperature. Snead[1],[2] reports that this swelling has been associated with lattice dilation from point defect formation [6], though recent modeling has suggested that small interstitial clusters may also impact swelling [7].

Comparisons by ORNL [1] of temperatures inferred from SiC measurements and thermocouples indicate that accuracies of approximately 20 °C are possible for ranges of 1 to 8 dpa and from 200 to at least 800 °C. Absolute temperature limits for resistivity techniques are typically stated as 150 °C (an amorphorous threshold) and 875 °C (due to recrystallization), but [2] suggests that electrical resistivity techniques may provide insights beyond these temperature limits.

Figure 1 shows the results of comparison evaluations performed by INL and ORNL on “mirrored” SiC temperature monitors irradiated in ORNL’s HFIR [8]. The peak irradiation temperature is identified when data extend beyond a band (see red dashed lines in Figure 1) corresponding to the maximum and minimum values measured at low temperatures and continue to exponentially increase (or decrease) with increasing temperature anneals.

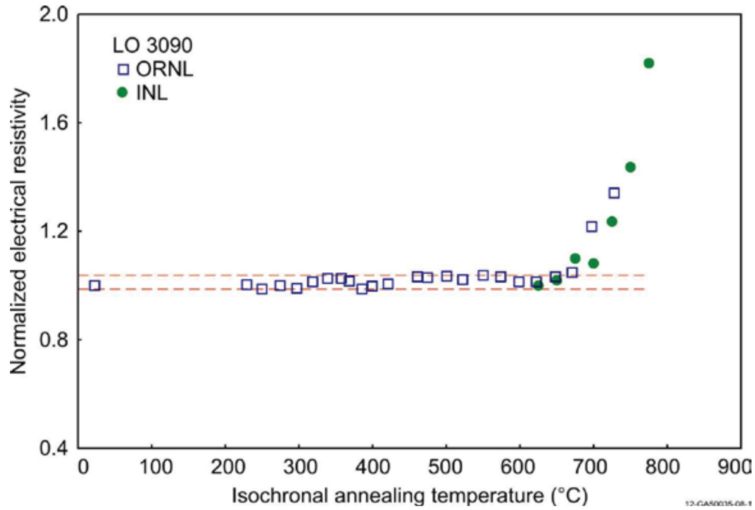


**Figure 1. INL and ORNL results for L03090 [8].**

There are several limitations associated with the use of SiC temperature monitors. As discussed above, temperatures are inferred by post-irradiation detection of changes in electrical resistivity associated with the stable defect population within SiC monitors that were incurred during irradiation. Higher accuracies are possible if the monitors are obtained from fully dense (3.2 g/cm$^3$), chemical vapor deposition (CVD) SiC and if the monitors are irradiated at a constant temperature. However, there are some limitations to this method as outlined for the following conditions [8]:

- *Irradiation temperatures rising during the latter part of irradiation*. SiC swelling saturates at low fluence. For damages greater than > 0.1 dpa, the increasing temperature will anneal out defects that occur at the lower irradiation temperature while creating stable defects at the higher temperature. When isochronal annealing is performed, lower temperature defects (to some extent) will have already been removed, and the recovery curve will be smeared to somewhat higher temperatures.
- *Irradiation temperatures decreasing during irradiation*. This decrease will lead to defects being created and frozen-in at the higher temperature, while continuing to create lower temperature defects. The isochronal anneal will then give an indication of the lowest irradiation temperature (in this case at the end of the irradiation period) and the recovery curve will be smeared because the annealing process will produce defects that remain at higher temperatures.
- *Upward or downward temperature spikes during irradiation.* Depending on the time at temperature, the effect will be to smear the recovery curve.

However, as noted above, if irradiation tests are conducted at or near the same temperature when the reactor is at power, none of these situations are of concern.

# 3 ATR CONFIGURATION

The ATR is a versatile tool for conducting nuclear reactor, nuclear physics, reactor fuel, and structural material irradiation experiments [9]. The ATR's maximum power rating is 250 $MW_{th}$ with a maximum unperturbed thermal neutron flux of $1x10^{15}$ n/cm$^2$-s and a maximum fast flux of $5x10^{14}$ n/cm$^2$-s.

As shown in Figure 2, the ATR core consists of 40 curved plate fuel elements in a serpentine arrangement around a 3 x 3 array of primary testing locations, including nine large high-intensity neutron flux traps. In addition to the nine large volume (up to 1.22 m long and up to 0.13 m diameter) high-intensity neutron flux traps, there are 66 irradiation positions inside the reactor core reflector tank and 34 low-flux irradiation positions in two capsule irradiation tanks outside the core.

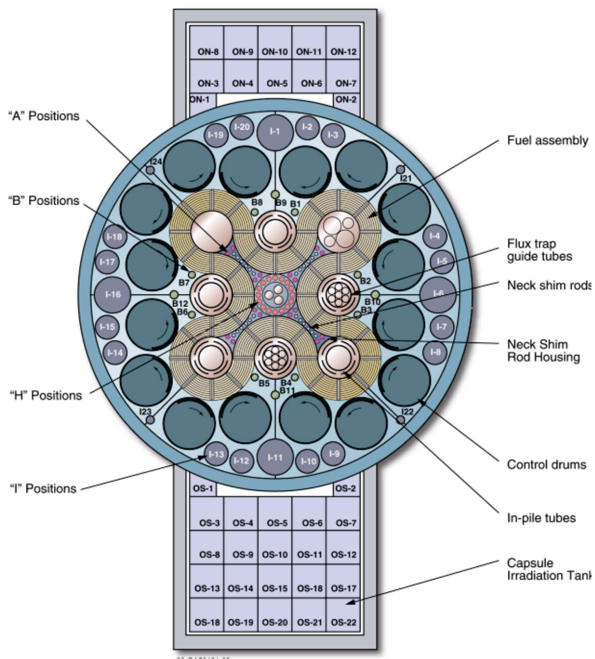


**Figure 2. ATR core cross section showing irradiation locations.**

ATR irradiation options and temperature sensors include:

- Static capsules that may contain several small samples or engineered components. Static capsule experiments may be sealed or may open to ATR primary coolant. Irradiation temperature may be selected (within limits) by providing a gas gap in the capsule with a known thermal conductance. Prior to the current effort, temperature monitoring for such locations was limited to sensors that detect peak temperature such as temperature activated paint spots or melt wires.
- Instrumented lead experiments that allow active control of experiments and data from test capsules during irradiation using core positions with instrumentation cables. In addition to

the sensors used in static capsules, these experiments can use thermocouples for real time temperature monitoring.

- Five of the nine ATR flux traps used for materials and fuels testing that are equipped with pressurized water loops (at the NW, N, SE, SW, and W locations) capable of replicating the conditions of a pressurized water reactor (PWR). These loops can operate at or above the standard temperatures and pressure of a commercial PWR power plant. Sensor options are the same as for an instrumented lead test.
- The HSIS or "rabbit" that enables insertion and removal of experiment specimens during ATR during operational cycles. Sensor options are the same as for a static capsule test.

## 4 EVALUATION OF SIC MONITORS AT THE HTTL

Specialized equipment was installed at INL's HTTL and specialized procedures were developed to ensure that accurate peak irradiation temperature measurements are inferred from SiC monitors irradiated at the ATR [10]. Key aspects of these procedures and this equipment are described below.

Figure 3 shows the equipment at INL's HTTL that has been configured to measure the electrical resistivity of SiC monitors after annealing. SiC monitors (approximately 0.5 to 2 mm x 0.5 to 2 mm x 10 to 25 mm) are heated to a temperature ranging from 200 to 800 ºC (where the annealing temperature range is selected to encompass peak irradiation temperatures predicted by thermal analyses for these specimens). The furnace used to heat these samples is placed under a ventilation hood located within the stainless steel enclosure.

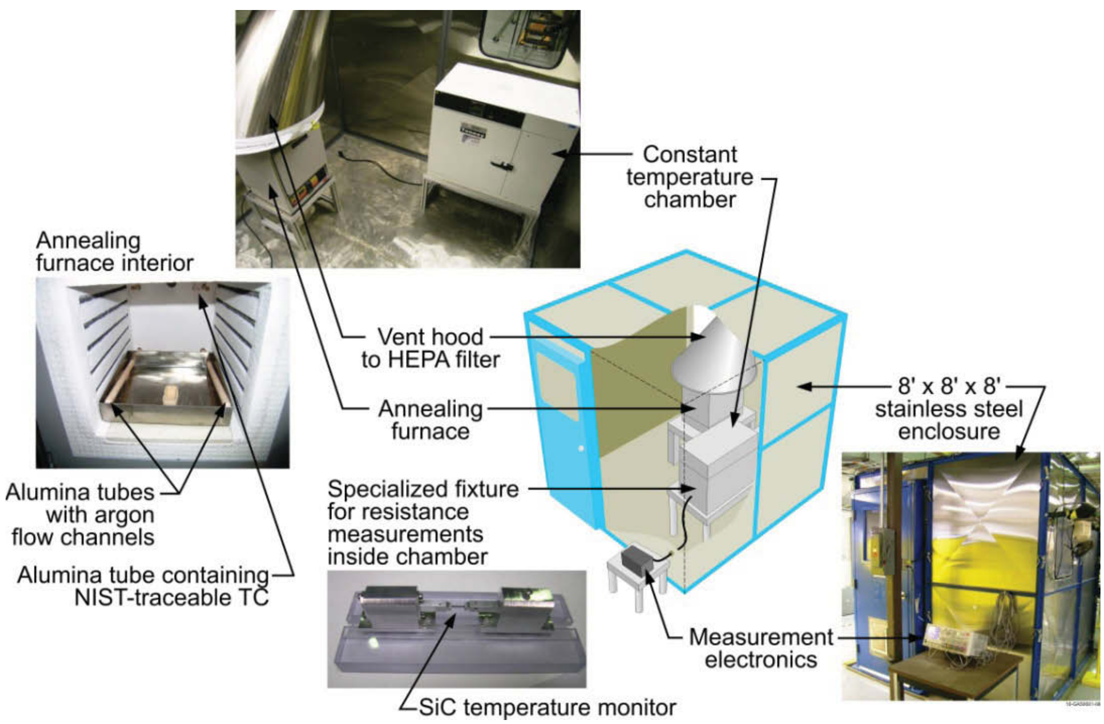


**Figure 3. Setup for annealing and measuring SiC temperature monitors**

Several modifications were made to the furnace in this setup to facilitate SiC temperature monitor annealing. Temperature accuracy in this furnace is verified using a National Institute of Standards and Technology (NIST) traceable thermocouple inserted into the furnace. Figure 4 shows the alumina tube that contains this thermocouple close to the top of the furnace. The outer two alumina tubes contain thermocouples that were provided by the furnace vendor for control and over-temperature protection.

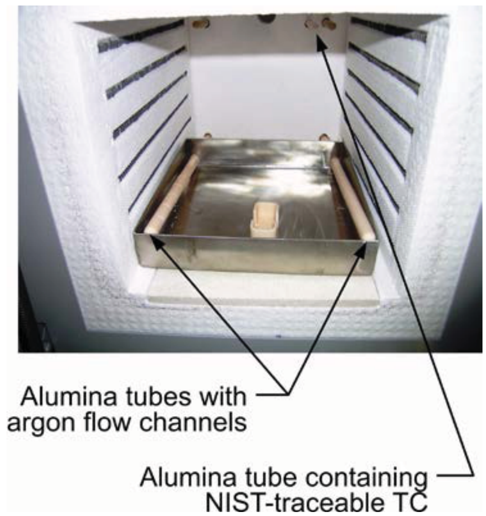


**Figure 4. Interior of annealing furnace.**

During heating, samples are placed in the alumina crucible (see Figure 4). Surface oxidation is removed from samples prior to resistivity measurement evaluations using a dilute hydrofluoric wash. Likewise, care must be taken to address surface silica formation if annealing temperatures exceed 600 °C in an air atmosphere. One option is to soak annealed samples in a dilute hydrofluoric wash. However, INL primarily relies on an alternate approach, which includes purging the annealing furnace with an inert gas to preclude silica formation during such tests (although the option of returning the SiC monitors to another location with a hydrofluoric wash is also available). Figure 4 shows a stainless steel pan with the alumina crucible and two alumina tubes with flow channels for the purge gas. During heating, samples are placed in this alumina crucible for 30 ± 2 minutes. To preclude surface silica formation if annealing temperatures exceed 600 °C in an air atmosphere, INL purges the annealing furnace with ultra-high purity (99.999%) argon gas using the two alumina tubes with flow channels. Argon is supplied to the furnace with a flowrate of a minimum of 2.7 liters/minute for a minimum of 30 minutes prior to SiC monitor insertion. To verify that silica formation has not occurred, INL evaluates a 'reference' unirradiated SiC monitor with each irradiated monitor after annealing. If the electrical resistivity of the 'reference' SiC monitor changes, both the 'reference' and the irradiated SiC monitors are treated with a dilute hydrofluoric wash.

After heating, the samples are placed in a constant environmental temperature chamber where current and voltage measurements are taken. This constant temperature environmental test chamber is used to ensure electrical measurements are taken within 0.2 °C of a predetermined temperature, typically set at 30 or 40°C. Electrical measurements must be taken to within 0.4 °C of the same temperature or it will adversely affect the accuracy of resistance values estimated for SiC temperature monitors.

Temperature stability within this furnace is verified by comparing furnace temperatures with the temperature from a calibrated thermocouple inserted into the constant temperature chamber. A high accuracy (9 digit) DC power analyzer is used to obtain electrical measurements. This power analyzer, which is located outside the stainless steel enclosure, is capable of applying constant voltages ranging from 1.5 mV to 50 V. For typical INL measurements, a constant voltage of approximately 20 V is applied, resulting in a microamp current flow that minimizes any unwanted effects from inadvertent resistance heating.

Figure 5 shows a close-up of the specialized fixture developed by INL for holding SiC monitors with rectangular or circular cross sections with a sketch illustrating the process used to make these

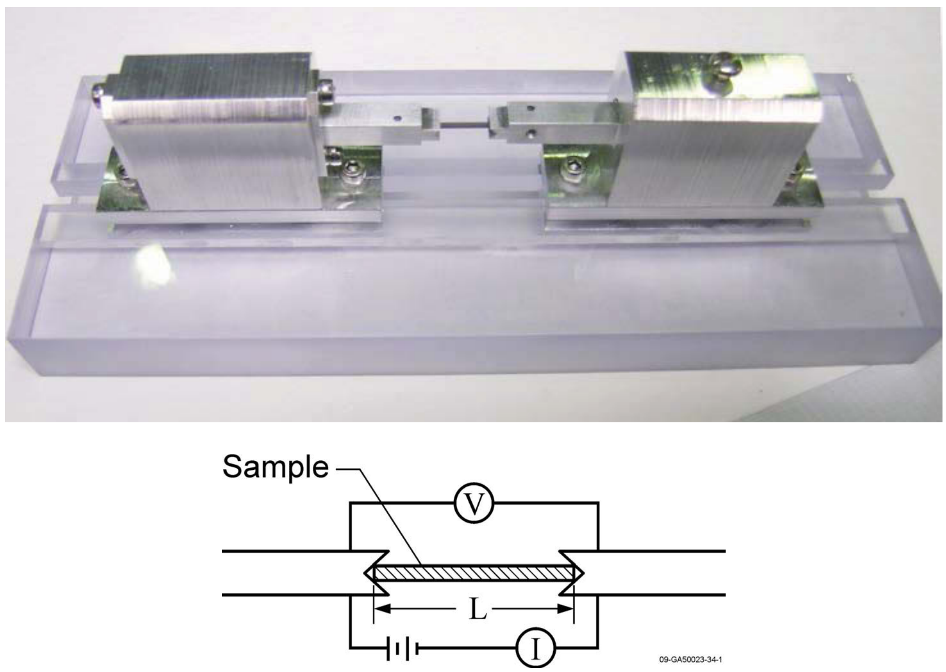


**Figure 5. Close-up of INL fixturing for making electrical measurement.**

measurements. As indicated in this figure, INL fixtures contact the ends of the SiC monitor, thus ensuring that electrical measurements are taken on the same portion of the sample during each measurement. A four point probe technique is used with the four points connected to the sample through spring-loaded angled copper electrodes that hold the SiC temperature monitor in place. Two sizes of angled electrodes are available at INL to accommodate different sizes of rectangular-shaped SiC monitors. In addition, conical shaped electrodes are available for rod-shaped SiC monitors.

Current and voltage are provided to the sample through wires that are threaded through the holes in the electrodes. The power analyzer applies a constant voltage to the sample. Then, the sample electrical resistivity is calculated using the measured voltage and current as shown in Figure 5.

The resistivity, R’, in ohm-m, is calculated using the equation $R' = (VA)/(IL)$

where

V = measured voltage (volts)

I = current resulting from the power supply voltage (amps)

L = sample length (m), and

A = sample cross-sectional area ($m^2$).

The ohmic nature of the sample is first evaluated (e.g., the measured current changes in direct proportion to the applied voltage). Then, a voltage is selected that minimizes resistance heating of the sample. This is done by applying a specified voltage and noting whether there is a rapid change of the measured current. In addition, the voltage is selected that yields a measurable current for this power analyzer.

# 5 EVALUATION OF SIC MONITORS

The performance of advanced nuclear systems relies on the performance of the materials used for fuel, cladding, and other components. In many proposed advanced systems, the reactor design pushes the temperature and the total radiation dose higher than typically seen in a current light water reactor. The objective of the University of Wisconsin Pilot Project [4] is to irradiate materials of interest for advanced reactor applications at a variety of temperatures (nominally 300 °C, 400 °C, 500 °C, and 700 °C) and total dose accumulations (nominally 3 dpa and 6 dpa). Peak irradiation temperatures during testing are inferred via post-irradiation examination of SiC temperature monitors. The first in a series of irradiation tests began in September 2008, and the first two SiC temperature monitors were evaluated at the HTTL in December 2012. These monitors were irradiated to have a target dose of 3 dpa and were fabricated from high density (3.2 $g/cm^3$) CVD SiC with a nominal size of 12.5 mm x 1.0 mm x 0.75 mm. Table 1 lists the expected peak irradiation temperature range for each monitor based on preliminary thermal analysis. The basket containing these monitors malfunctioned during the irradiation test affecting the in-core location by approximately 12 mm. Post irradiation calculations are planned to reduce uncertainties in these estimated temperatures as a result of the basket movement.

**Table 1. SiC temperature monitors tested.**

| Monitor Identification | Expected Peak Irradiation Temperature Range (°C) |
|---|---|
| 05R4-02-A KG1403 (300 LO) | 287 - 343 |
| 05R4-01-A KG1415 (400 LO B) | 411 - 442 |

This section discusses the evaluation of the SiC monitors and presents the results. As discussed in Section 4, testing was conducted in accordance with Reference [10]. Sections 5.1 and 5.2 present the data collected for each monitor and provide interpretation of the data.

### 5.1 SiC Temperature Monitor 300 LO

Prior to heating, the ohmic response for the 300 LO monitor was tested and found to be linear. These data were used to select a test voltage that would result in minimal heating of the SiC monitor during electrical resistance testing and remain within the range of the test instrumentation. For this testing, an applied voltage of 2V was selected.

Figure 6 presents the electrical resistivity data taken at each isochronal annealing temperature. As discussed in Section 2, the peak irradiation temperature using an electrical resistivity technique is the point where the resistivity begins, and consistently remains, above the error band. The error band bounds the data and is represented by the red dotted lines shown in Figure 6. Using this method, the first data point to consistently remain above the error band occurred after an anneal at 295 °C. This temperature falls within the planned irradiation temperature range of 287 to 343 °C. As noted in Section 2, the uncertainty using this technique is generally accepted to be 20 °C (although it can increase depending on selected temperature intervals).

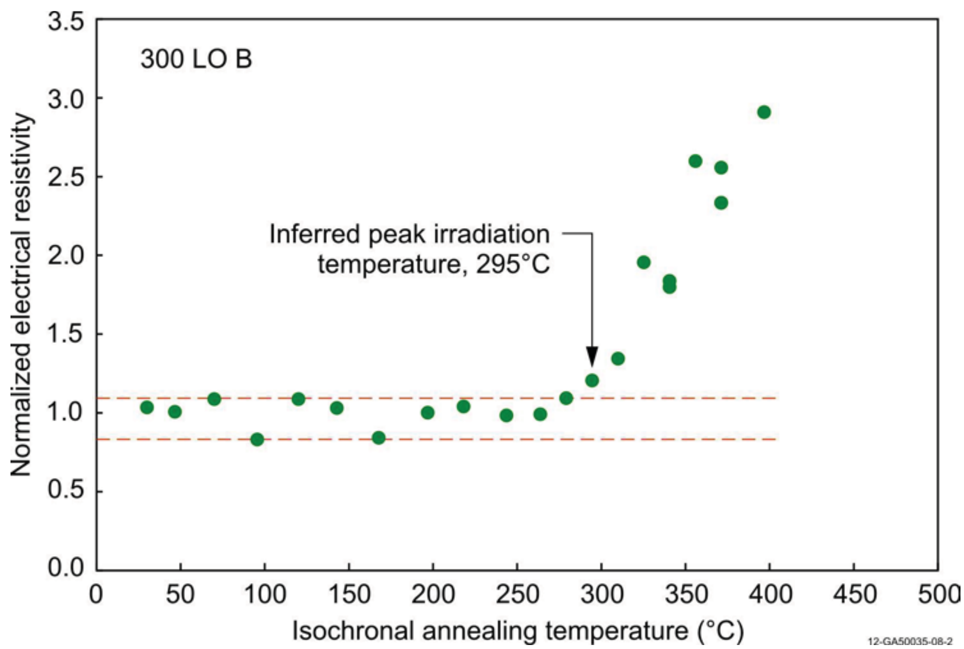


**Figure 6. Electrical resistivity data for the 300 LO monitor.**

### 5.2 SiC Temperature Monitor 400 LO B

As discussed in Section 2, if SiC temperature monitor evaluations consider samples with peak annealing temperatures greater than 400°C, the INL procedure for evaluating SiC temperature monitors [10] requires the use of a reference unirradiated SiC monitor and the use of an argon atmosphere during annealing. Hence the 400 LO B evaluations included a reference sample. For the 400 LO B and reference sample, an ohmic response evaluation indicated that the appropriate applied voltage would be 2V and 20V, respectively.

Figure 7 presents the electrical resistivity data taken after each isochronal annealing for the reference monitor as well as for the 400 LO B monitor. Oxidation of the reference monitor during the annealing process would have resulted in a substantial increase in resistivity for the reference monitor. However, as shown in Figure 7, no significant increase in electrical resistivity was observed.

Figure 7 also presents the electrical resistivity data taken at each isochronal annealing temperature for the 400 LO B monitor. As shown in Figure 7, the first data point to consistently remain above the error band was obtained after an annealing temperature of 294 °C. This temperature is considerably lower than the planned irradiation temperature range of 411 to 442 °C. Because of this unexpected result, the electrical resistivity measurement was repeated several times. Each data point taken consistently remained above the error band. Hence, measurements indicated that the peak irradiation temperature is 294 °C with the uncertainty estimated as ±25 °C. Note that the larger uncertainty is because larger temperature increments (25 °C) were selected when significant increases in resistivity were observed. Smaller increments in annealing temperatures had not been planned until temperatures were closer to the expected values of 411 to 442 °C.

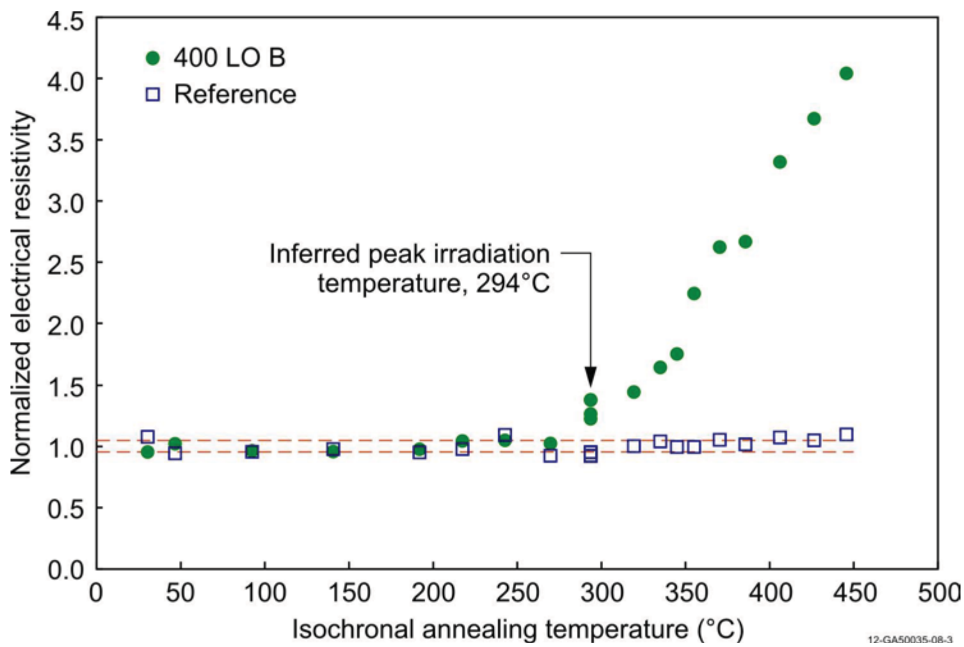


**Figure 7. Electrical resistivity data for the 400 LO B and reference monitor.**

As noted in Section 2, there are several limitations associated with the use of SiC temperature monitors that limit their accuracy. For example, if the irradiation temperature was lower for a time period after the peak irradiation temperature occurred, lower peak irradiation temperatures will be inferred from SiC resistivity measurements. Obviously, the reported changes in test geometry (basket malfunction) or other boundary condition changes also affect the peak irradiation temperature. Post irradiation thermal analyses are planned to help understand the peak irradiation temperature measured by the 400 LO B monitor.

# 6 SUMMARY

Evaluations of the first two SiC monitors from the University of Wisconsin Pilot Project [4] have been completed. Table 2 summarizes peak irradiation temperatures inferred from electrical resistivity measurements of these monitors. The peak irradiation temperature inferred for the 300 LO monitor was 295 ± 20 °C. This temperature was within the expected peak irradiation temperature range. The peak irradiation temperature inferred for the 400 LO B monitor was 294 ± 25 °C. Post irradiation thermal analysis may prove helpful in explaining the disparity between the peak irradiation temperature (294 °C) and the expected irradiation temperature range (411- 442 °C). Data obtained from an unirradiated reference SiC monitor did not indicate that any oxidation occurred during annealing.

**Table 2. Test results for the SiC monitors tested.**

| Monitor Identification | Inferred Peak Irradiation Temperature (°C) | Error (°C) | Expected Peak Irradiation Temperature Range (°C) |
|---|---|---|---|
| 05R4-02-A KG1403 (300 LO) | 295 | ±20 | 278 - 343 |
| 05R4-01-A KG1415 (400 LO B) | 294 | ±25 | 411- 442 |

# 7 ACKNOWLEDGMENTS

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# 8 REFERENCES

1. L L. Snead, A. M. Williams, and A. L. Qualls, "Revisiting the use of SiC as a post irradiation temperature monitor," *Effects of Radiation on Materials*, *ASTM STP 1447*, M L. Grossbeck, Ed, ASTM International, West Conshohocken, PA, 2003.

2. L. Snead, "Revisiting the use of SiC as a post irradiation temperature monitor," *Presented at the International Symposium on Materials Test Reactors,* Idaho Falls, ID, September 28, 2009.

3. Davis, K.L., B. Chase, T. Unruh, D. Knudson, and J. Rempe, "Evaluations of University of Wisconsin Silicon Carbide Temperature Monitors 300 LO and 400 LO B," INL/EXT-11-24226, Idaho National Laboratory, December 2011.

4. MacLean, H.J, and K. Sridharan, T. A. Hyde, “Irradiation Test Plan for the ATR National Scientific User Facility - University of Wisconsin Pilot Project,” PNL-2784, Revision 1, Idaho National Laboratory, July 21, 2008.

5. N. F. Pravdyuk, V. A. Nikolaenko, V. I. Kapuchin, V.N. Kusnetsov, in: Ed. D. J. Littler, *Proceedings of the Properties of Reactor Materials and the Effects of Radiation Damage,* Butterworths, 1962, p. 57.

6. M. Balarin, “Zür Temperaturabhangigkeit der Strahlensattigung in SiC,” *Physica Status Solidi*, 11, Issue 1, (1965), K67.

7. H. Huang and N. Ghoniem, “A swelling model for stoichiometric SiC at temperatures below 1000 degrees C under neutron irradiation,” *Journal of Nuclear Materials*, 250, Issues 2-3, (December 1997), p. 192.

8. J. L. Rempe, K. G. Condie, D. L. Knudson, and L. L. Snead, “Comparison Measurements of Silicon Carbide Temperature Monitors,” *IEEE Transactions on Nuclear Science*, **57**, No. 3, June 2010.

9. “FY2009 Advanced Test Reactor National Scientific User Facility User’s Guide,” INL/EXT-08-14709, Idaho National Laboratory, 2008.

10. Rempe, J.L. and K.G. Condie, D. L. Knudson, “Silicon Carbide Temperature Monitor Evaluation,” PLN-3473, Revision 0, Idaho National Laboratory, May14, 2010.