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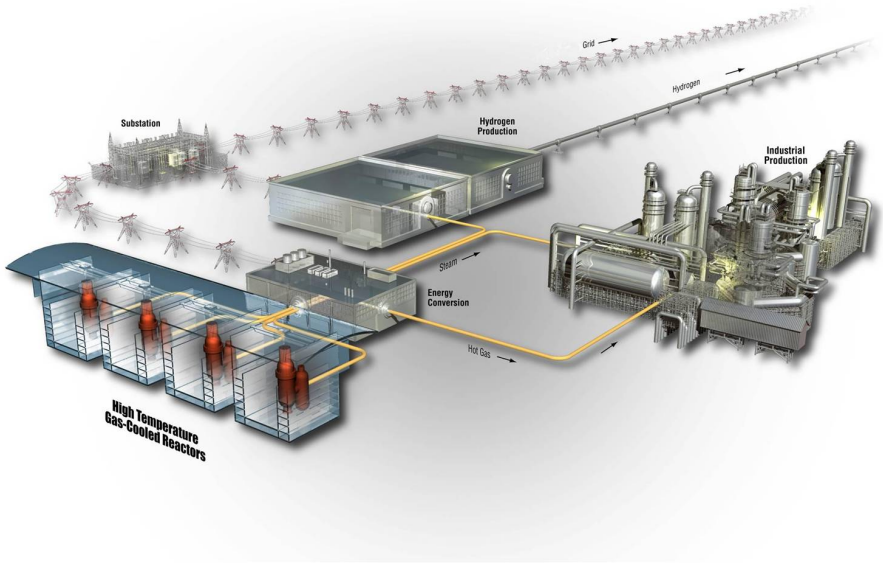
Plan

AGC-4 Graphite Specimen Post-Irradiation Characterization Plan



W. David Swank, William Windes and David Rohrbaugh

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Approved by:

08/26/2020

William E. Windes
ART Graphite R&D Lead

Date

Michael Davenport see eCR 681358

Michael Davenport
ART Project Management

Date

Michelle Sharp

08/27/2020

Michelle Sharp
Quality Assurance

Date

REVISION LOG

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SUMMARY

This characterization plan describes the thermal, physical, and mechanical measurement techniques that will be used to characterize graphite samples being tested in the fourth Advanced Graphite Creep experiment (AGC-4). Instruments, fixtures, and methods are currently in place for both pre- and post-irradiation material property measurements of bulk density, thermal diffusivity, coefficient of thermal expansion, elastic modulus, and electrical resistivity. Post-irradiation testing procedures used to characterize the samples are described and discussed in the plan. Where they exist, American Society for Testing and Materials (ASTM) international testing standards will apply to the tests. Any departure from ASTM international testing standards or the approved laboratory procedures are documented within this characterization plan. Deviations that occur during testing will be documented in data reports.

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TABLES

ACRONYMS

AGC	Advanced Graphite Creep
ART	Advanced Reactor Technology
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials International
ATR	Advanced Test Reactor
CCL	Carbon Characterization Laboratory
COV	coefficient of variation
CTE	coefficient of thermal expansion
HDG	High Dose Graphite
HTR	high-temperature reactor
INL	Idaho National Laboratory
IRC	INL Research Center
LFA	laser flash apparatus
NDMAS	NGNP Data Management and Analysis System
NGNP	Next Generation Nuclear Plant
PIE	post-irradiation examination
R&D	research and development

1. INTRODUCTION

The Next Generation Nuclear Plant (NGNP) Graphite Research and Development (R&D) program has been combined with other nuclear material research programs to form the Advanced Reactor Technology (ART) materials program. Similar to the NGNP Graphite program, the ART Graphite R&D program is investigating and qualifying graphite for nuclear applications. Graphite has been used effectively as a structural and moderator material in both research and commercial high-temperature gas-cooled, molten-salt-cooled, and other reactor designs. While the knowledge and materials necessary to produce nuclear-grade graphite are generally understood, historical nuclear grades no longer exist. Therefore, new grades must be fabricated, characterized, and irradiated to demonstrate that current grades of graphite exhibit acceptable irradiated and nonirradiated properties, upon which the thermomechanical design of the structural graphite in a graphite core reactor design is based. All activities associated with graphite characterization are performed under PLN-2690, "Idaho National Laboratory Advanced Reactor Technologies Quality Assurance Program Plan." Further details on the R&D activities and associated rationale needed to qualify nuclear-grade graphite for use within a graphite reactor design have been documented in PLN-2497, "Graphite Technology Development Plan," and INL/EXT-05-00269, "NGNP Graphite Testing and Qualification Specimen Selection Strategy."

The Advanced Graphite Creep (AGC) experiment, consisting of six irradiation capsules, is generating irradiated graphite performance data for high-temperature reactor (HTR) operating conditions. The AGC experiment is designed to determine the changes to the specific material properties of thermal diffusivity, thermal expansion, elastic modulus, mechanical strength, irradiation-induced dimensional change rate, and irradiation creep for a wide variety of nuclear-grade graphite types. In 2018, the Department of Energy approved a major design change to the ART AGC experiment. It was determined that the AGC experiment should extend the neutron dose range from 0–7 dpa to a 0–15 dpa to be pertinent to current HTR designs. This new neutron dose increase will extend the current nuclear grades past turnaround dose levels and into the nonlinear tertiary-creep regime. To achieve this higher maximum dose level, it was decided to repurpose the last two irradiation capsules, AGC-5 and AGC-6, which were to be irradiated under very high-temperature reactor conditions of 1100°C. Under the new direction given by DOE in 2018, AGC-5 and AGC-6 capsules will be used to irradiate previously exposed specimens from AGC-2, AGC-3, and AGC-4, once they have undergone post-irradiation examination (PIE). AGC-5 will be renamed as High Dose Graphite (HDG)-1 and will re-irradiate AGC-2 specimens at a nominal irradiation temperature of 600°C. AGC-6 will be renamed as HDG-2 and will re-irradiate selected specimens from AGC-3 and AGC-4 at a nominal irradiation temperature of 800°C. No graphite specimens will be irradiated at temperatures of 1100°C. Once irradiation is complete in HDG-1 and HDG-2, all specimens will undergo additional PIE testing to determine the effects of this higher dose level on material properties.

All six AGC capsules in the experiment are irradiated in the Advanced Test Reactor (ATR). After irradiation, the AGC capsules are cooled in the ATR canal, sized for shipment, and shipped to the Materials and Fuels Complex, where they are disassembled in the Hot Fuel Examination Facility. During disassembly, the metallic capsules are machined open, and the individual samples are removed from the interior graphite body of the capsule. Samples removed from the capsule are loaded in a shipping drum and shipped to the Idaho National Laboratory (INL) Research Center (IRC) for initial PIE and storage for any future testing at the Carbon Characterization Laboratory (CCL).

The CCL is located in Laboratories C-19 and C-20 of IRC. It was specifically designed to support graphite and ceramic composite research and development activities (INL/EXT-11-22148). The CCL is designed to characterize and test low-activated irradiated materials, such as high-purity graphite, carbon-carbon composites, and silicon-carbide composite materials. The laboratory is fully capable of

characterizing material properties for both irradiated and nonirradiated materials. All test specimens from each of the six capsules will be processed through the CCL to visually inspect each sample, perform initial dimensional changes, and repackage the samples for shielded storage in the irradiated graphite vault located in Laboratory C-19, as shown in Figure 1 and Figure 2.

Figure 1. The CCL glovebox used to visually inspect graphite samples, perform initial dimensional measurement, and repackage samples for storage in the irradiated graphite vault in Laboratory C-19.

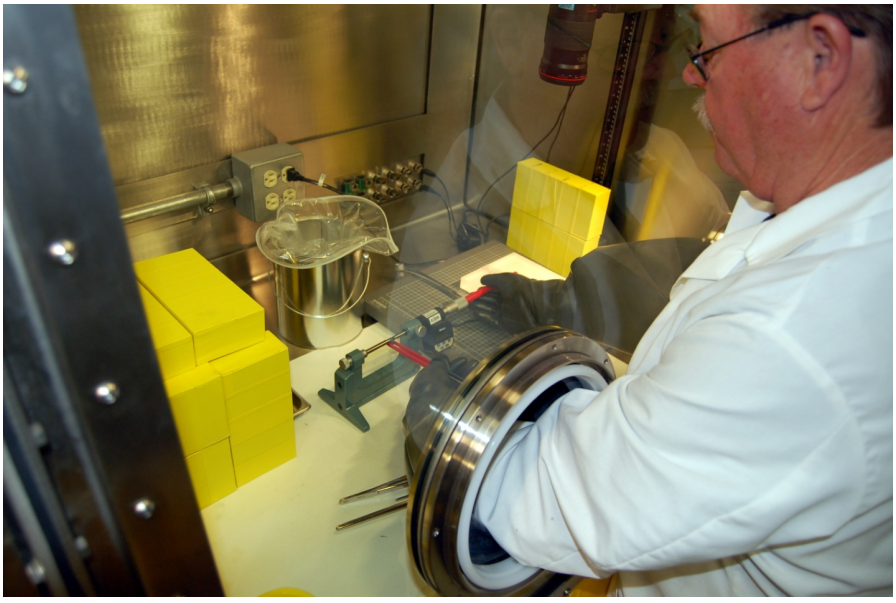


Figure 2. Dimensional measurement and transfer of samples to plastic storage containers.

This characterization plan describes the thermal, physical, and mechanical measurement techniques and methods that will be used to characterize the different graphite types being tested in AGC experiments. It is intended to meet the requirements of INL MCP-1380, "Research and Development Test Control," and those of American Society of Mechanical Engineers Quality Assurance Requirements for Nuclear Applications ASME-NQA-1-2008/1a-2009, "Quality Assurance Requirements for Nuclear Facility Applications." Instruments, fixtures, and methods are currently in place for pre- and post-irradiation material property measurements of bulk density, thermal diffusivity, coefficient of thermal expansion (CTE), elastic modulus, and electrical resistivity. Table 1 lists the instruments, material properties measured, and the following applicable American Society for Testing and Materials International (ASTM) standard against which each measurement will be performed:

- ASTM C559-16, "Standard Test Method for Bulk Density by Physical Measurements of Manufactured Carbon and Graphite Articles"
- ASTM C611-98(2016), "Standard Test Method for Electrical Resistivity of Manufactured Carbon and Graphite Articles at Room Temperature"
- ASTM C747-16(Reapproved 2010), "Standard Test Method for Moduli of Elasticity and Fundamental Frequencies of Carbon and Graphite Materials by Sonic Resonance"
- ASTM C769-98(2005), "Standard Test Method for Sonic Velocity in Manufactured Carbon and Graphite Material for Use in Obtaining an Approximate Young's Modulus"
- ASTM E228-17, "Standard Test Method for Linear Thermal Expansion of Solid Materials with a Push Rod Dilatometer"
- ASTM E1461-13, "Standard Test Method for Thermal Diffusivity by the Flash Method."

Table 1. CCL measurement and test equipment.

Measurement	Standard	Instrumentation	Calibration Method	Result
Physical Dimensions and Mass	ASTM C559-16	Mitutoyo Micrometer 121-155: INL ID: 725884 INL ID: 727312 Mitutoyo Caliper CD-6″ CSX: INL ID: 725813 INL ID: 726607 INL ID: 727194 Sartorius Scale ME235P: INL ID: 412642 INL ID: 415907	INL Standards and Calibration Laboratory	Bulk density
Fundamental Frequency	ASTM C747-16	J. W. Lemmens Grindosonic: INL ID: 412850	No calibration required per instrument manufacturer	Elastic modulus
Sonic Velocity	ASTM C769-98(2005)	Olympus NDT Sq. Wave Pulser/Receiver 5077PR: INL ID: 728024 National Instruments Digitizer: USB 5133 INL ID: 726725 INL ID: 415868	INL Standards and Calibration Laboratory	Young's modulus, Shear modulus, Poisson ratio
4-Point Electrical Resistivity	ASTM C611-98(2016)	Kiethly 6220 Precision Current Source: INL ID: 725865 INL ID: 727290 Kiethly 2182A Nano Voltmeter: INL ID: 725866 INL ID: 727289	INL Standards and Calibration Laboratory	Electrical resistivity
Laser Flash Diffusivity	ASTM E1461-13	Netzsch Laser Flash Apparatus 457 2 ea.: INL ID: 412855 INL ID: 412864	Calibration by user per manufacturer's instructions	Thermal diffusivity
Push Rod Dilatometry	ASTM E228-17	Netzsch DIL 402 C 2 ea.: INL ID: 412860 INL ID: 412861	Calibration by user per manufacturer's instructions	CTE
Environmental Monitoring	ALL	Visala Pressure, Humidity and Temperature PTU301: INL ID: 726912 INL ID: 727884 INL ID: 727502	INL Standards and Calibration Laboratory	Laboratory environmental conditions

Note: *All ASTM standards are reapproved every five years. During this review, the technical content of the standard is confirmed to still be valid. In some cases, this process of reapproval will necessitate a more in-depth revision. When a new revision takes place, the publication year is changed. Over the long*

duration of the full ART AGC irradiation experiment matrix, the ASTM standards followed will be reapproved and revised. These standards are reviewed for technical equivalency prior to each publication of an AGC capsule characterization plan. This makes it possible to compare and analyze data across the full experiment matrix. When a revision is technically equivalent to its predecessor, the new version is used and referenced throughout the full test (e.g., characterization plan, preirradiation analysis, and post-irradiation analysis). If the revision is not technically equivalent, the previous version will continue to be followed and referenced.

2. GENERAL PROVISIONS

The objective of the AGC experiments is to determine the material property changes induced in nuclear-grade graphite during exposure to a neutron environment. The approach is to perform extensive preirradiation characterization testing on each sample before exposing the graphite to neutron doses. After irradiation, the same characterization tests will be performed on the samples to ascertain the quantitative changes to the material properties of the nuclear-grade graphite.

Irradiation of the fourth AGC experiment (AGC-4) was completed in January 2020. A complete description of the irradiation parameters will follow in a separate report. This characterization plan provides details on how the samples are to be tested and characterized, which ASTM tests are to be used, which versions of the ASTM tests are to be used, and whether these methods and techniques are alterations of the original test standard. Finally, the plan describes how the data generated from the tests will be recorded and retained within the ART Graphite R&D project.

All work will be performed in accordance with INL LWP-20000, “Conduct of Research,” and INL Laboratory Instruction (LI)-709, “Irradiated Graphite Characterization,” is currently in place to govern irradiated graphite measurements.

All records designated in implementing documents as quality assurance records will be controlled in accordance with PLN-4653, “INL Records Management Plan.”

2.1 Specimen Description and Preparation

All specimens are 0.491 inch (12.47 mm) in diameter, with the creep specimens being 25.4 mm long and the piggyback specimens being 6 mm long. Details of how specimens were cut from the graphite blocks are contained in INL Drawing 778033, Rev. 1, “ATR Advanced Graphite Capsule (AGC) AGC-4 Graphite Specimen Cut-Out Diagrams.”

The AGC-4 specimens have been characterized per PLN-4239, “AGC-4 Graphite Specimen Preirradiation Characterization Plan,” prior to irradiation. This plan describes the thermal, physical, and mechanical measurement techniques and methods that were used to characterize the different graphite types being tested in the AGC-4 capsule. Described within the plan are the instruments, fixtures, and methods used for preirradiation material property measurements of bulk density, thermal diffusivity, CTE, elastic modulus, and electrical resistivity. The results of the preirradiation measurements, along with details of the graphite specimen grades and their position in the capsule/reactor, can be found in INL/EXT-16-38044, “AGC-4 Graphite Preirradiation Data Analysis Report.”

Following irradiation, specimens are unloaded from the AGC capsule and separated. Each specimen’s laser identification is matched to a barcoded snap-cap container, and the specimen placed in that container for storage. It should be noted that irradiated samples are not washed prior to characterization measurements.

2.2 Data Collection

Significant improvements in specimen test fixtures and data acquisition have been made specifically to support AGC specimen measurements. The equipment used to measure the material properties listed in Table 1 are segregated into individual stations that consist of the instrumentation necessary to make the measurement, a computer for automated data acquisition, and a barcode reader.

The barcode of the individual specimen container is read, and the file for that specimen is automatically opened for data input prior to each measurement. Associated with each measurement type is a unique laboratory notebook maintained to INL procedure MCP-2875, "Maintaining Laboratory Notebooks," and "INL Advanced Reactor Technologies Quality Assurance Program Plan" PLN-2690. Accepted data will be stored in the NGNP Data Management and Analysis System (NDMAS), a satellite file location for the ART Graphite R&D program. Data in a standardized Excel file format will be transmitted to the NDMAS using Form 250.01, "Data Management and Analysis Transmittal," per PLN-2709, "Nuclear Data Management and Analysis System Plan."

In addition to laboratory notebooks, the specific measuring instruments are networked to a server computer where the measurement data is automatically stored. This has been implemented in the INL CCL, where custom LabVIEW software was written to facilitate automated data acquisition. This software is comprised of five main programs: manufacturer's data, physical and dimensional measurements, electrical resistivity measurements, sonic resonance (fundamental frequency) measurements, and sonic velocity measurements. These five programs acquire data from instrumentation or user input and record the results in an Excel spreadsheet located on a server computer. In the case of thermal expansion and thermal diffusivity measurements, two other LabVIEW programs were written to parse vendor software acquired data into Excel spreadsheets. Form 562.41, "Software Management Plan and Life Cycle Documentation for R&D Activities," and LWP-13620, "Managing Information Technology Assets," are currently used to govern the development, accuracy, and configuration control of this software.

The measurements are made in the following sequence:

1. Initial dimensional measurements and photographs—all specimens
2. Specimens dried at 130°C for a minimum of 2 hours and held in a desiccator—all specimens
3. Mass and dimensional measurements—all specimens
4. Electrical resistivity—creep specimens
5. Elastic modulus by sonic resonance—creep specimens
6. Elastic modulus by measurement of sonic velocity—creep specimens
7. Thermal diffusivity—piggyback specimens
8. CTE up to irradiation temperature—creep specimens
9. CTE up to 1000°C—creep specimens
10. Postcharacterization of mass and dimensional measurements—all specimens.

2.3 Personnel and Training

Personnel who perform measurements identified in this plan are qualified in accordance with PDD-1300 "Quality Assurance Program Description". Their ability to adequately perform measurements described in this plan is demonstrated by instrument manufacturers' training and certification and/or the

performance of an instrument/measurement operational validation. Personnel qualifications are reviewed by the graphite research and development lead and documented in laboratory notebooks.

2.4 Variations, Exceptions, and Discrepancies

There are several variations, exceptions, and discrepancies that may occur. The first is a known departure from the applicable ASTM standard. These departures are typically related to geometrical constraints. All currently known departures or exceptions taken to the ASTM standard are described in detail in Section 3 of this plan. Any departure not captured in this plan will be recorded in laboratory notebooks associated with the measurement. In most cases, the effects of the exception or departure from the ASTM method on the measured value are not well understood. When possible, sensitivity studies will be performed and documented in laboratory notebooks to understand the impact of these exceptions and departures.

It is likely that the ASTM standards and/or test methods will be revised and improved during the 10+ year AGC experiment cycle. Each revision or development will be evaluated for how it could impact future measurements and their consistency with measurements made under previous revisions or techniques. A programmatic determination will be made whether to continue with the current version of the ASTM/method or use the updated version.

While measurements are being made, it is possible that something out of the ordinary may occur. Any unusual event that occurs during a measurement will be documented in the laboratory notebook associated with that specific measurement and duly noted within the database associated with the data generated for this program. The principal investigator will be notified of the event and then determine what impact it has on the data and document the significance of the result in the laboratory notebook.

2.5 Calibration and Functional Validation

The measurement protocol consists of calibration, functional validation, and data acquisition. Functional validations established for each measurement in collaboration with the instrument manufacturer are performed periodically to ensure that accurate and consistent data are acquired. All validations are performed on traceable standards and documented in retrievable laboratory notebooks associated with each measurement. In the event that an instrument functional validation fails, the reason for the failure is investigated and resolved prior to that measurement being used for further characterization. Upon resolution, a determination is made as to the impact the failure might have had on data taken prior to the failure and back to the last valid measurement. If the data captured during this interval is suspect, the impacted data is evaluated for credibility.

LWP-13455, “Control of Measuring and Test Equipment,” is followed for calibration standards, methods, and frequencies that have been established for each measurement. Where it is not possible to use the INL Standards and Calibration Laboratory, calibration by user procedures are established based on ASTM standards and manufacturers’ instructions and performed against international standards. These procedures are documented in laboratory notebooks associated with each measurement.

3. TEST METHODS

Before any measurements are made, specimen numbers and basic information about each type of graphite are entered into the manufacturer’s data program. A typical basic information interface for capturing this data is shown in Figure 3. Once basic information about the graphite type has been recorded, it is automatically saved to an Excel spreadsheet file, and the individual specimen numbers are entered using a barcode reader, as shown in Figure 4. Following the initial input of general information, individual material properties are measured, starting with mass and dimensional measurements for determining bulk density.

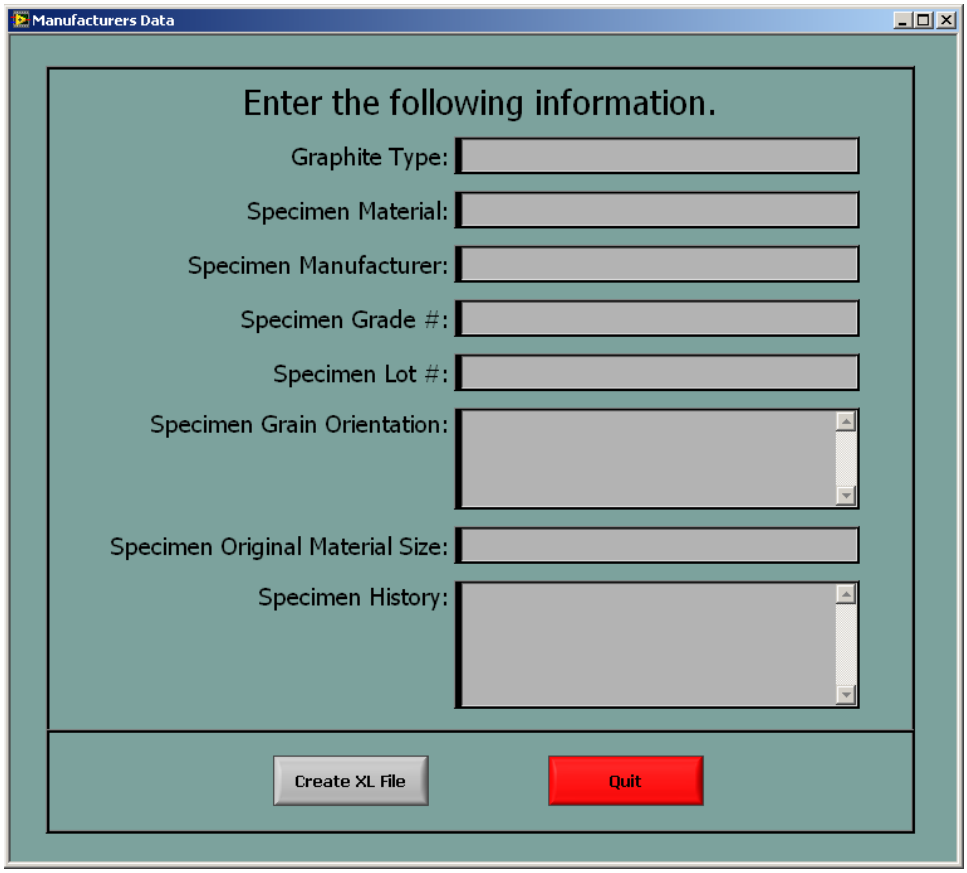
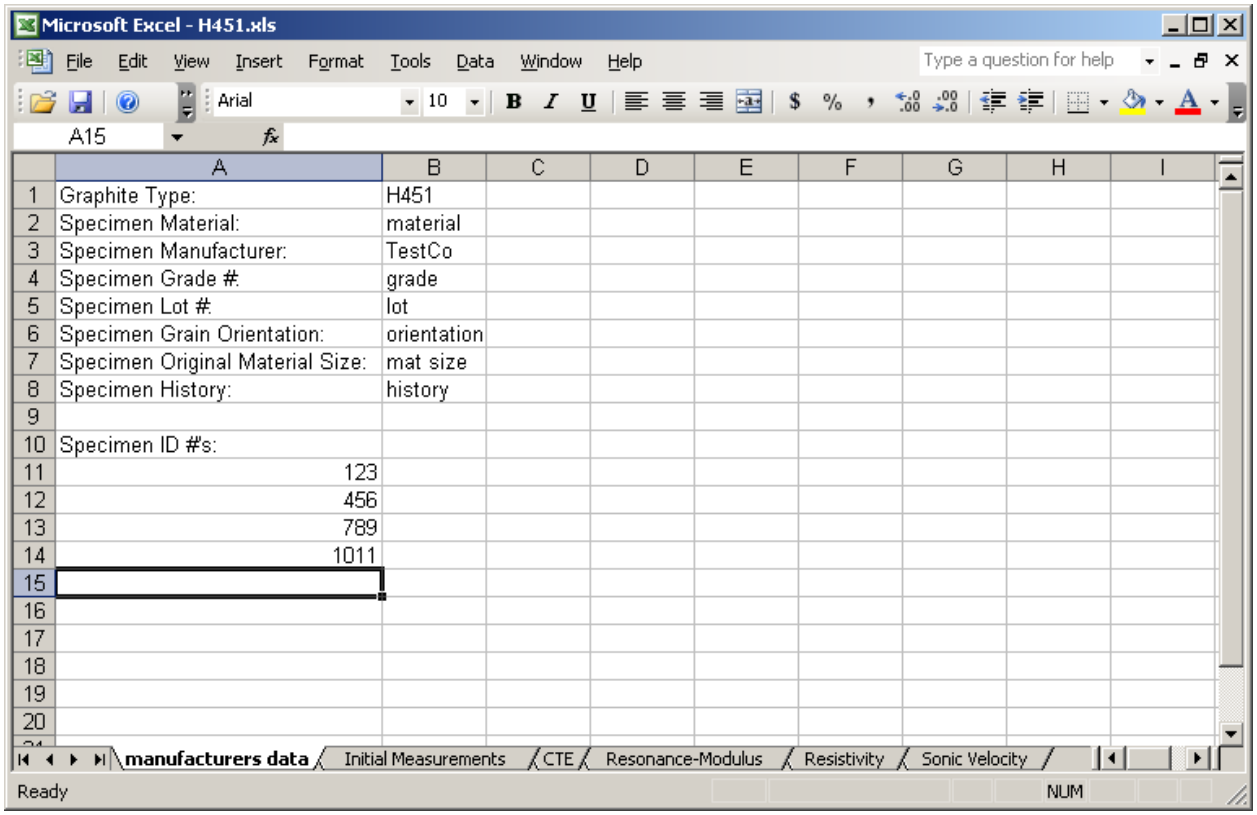


Figure 3. Manufacturer's data user interface screen.



	A	B
1	Graphite Type:	H451
2	Specimen Material:	material
3	Specimen Manufacturer:	TestCo
4	Specimen Grade #:	grade
5	Specimen Lot #:	lot
6	Specimen Grain Orientation:	orientation
7	Specimen Original Material Size:	mat size
8	Specimen History:	history
9		
10	Specimen ID #s:	
11	123	
12	456	
13	789	
14	1011	
15		
16		
17		
18		
19		
20		

Figure 4. Excel data spreadsheet example of the “manufacturer’s data” tab.

3.1 Mass and Dimensional Measurements

Dimensional change is one of the key parameters affecting the performance of graphite in a neutron environment. Determining the volumetric and linear dimension as a function of temperature and radiological dose will be necessary to understand critical performance measures, such as dimensional change turnaround, irradiation creep, and internal stresses imposed upon graphite components. Dimensional and mass measurements are performed to ASTM Standard ASTM C559-16, “Standard Test Method for Bulk Density by Physical Measurements of Manufactured Carbon and Graphite Articles,” which describes in detail the procedure for making dimensional measurements and calculating bulk density. Dimensional measurements of the specimen radius and length are made with INL-calibrated micrometers and calipers. The mass is measured using an INL-calibrated electronic balance. Figure 5 shows the dimensional measurement station.

The physical and dimensional measurements program user interface is shown in Figure 6. Measurement values are transferred directly from the measurement tools into the LabVIEW software. Once the physical and dimensional measurements of the specimens are taken, the data are automatically written to the Excel spreadsheet under the “Initial Measurements” tab. These data are used to calculate bulk density and are available for other measurement calculations.

The manufacturer accuracy of the dial micrometers is stated to be 2 µm. This is a 0.008% accuracy on a 25.4-mm measurement. However, when evaluating the uncertainty of the density determination, other factors must be considered, such as the hardness of the material and the force with which the micrometer blade is engaged with the material, specimen temperature variation, technician skill, etc. These and other

factors were considered in a propagation of error analysis to arrive at an uncertainty of 0.08% with the measurement of the diameter being the largest contributor to the error.

Figure 5. Dimensional measurement station.

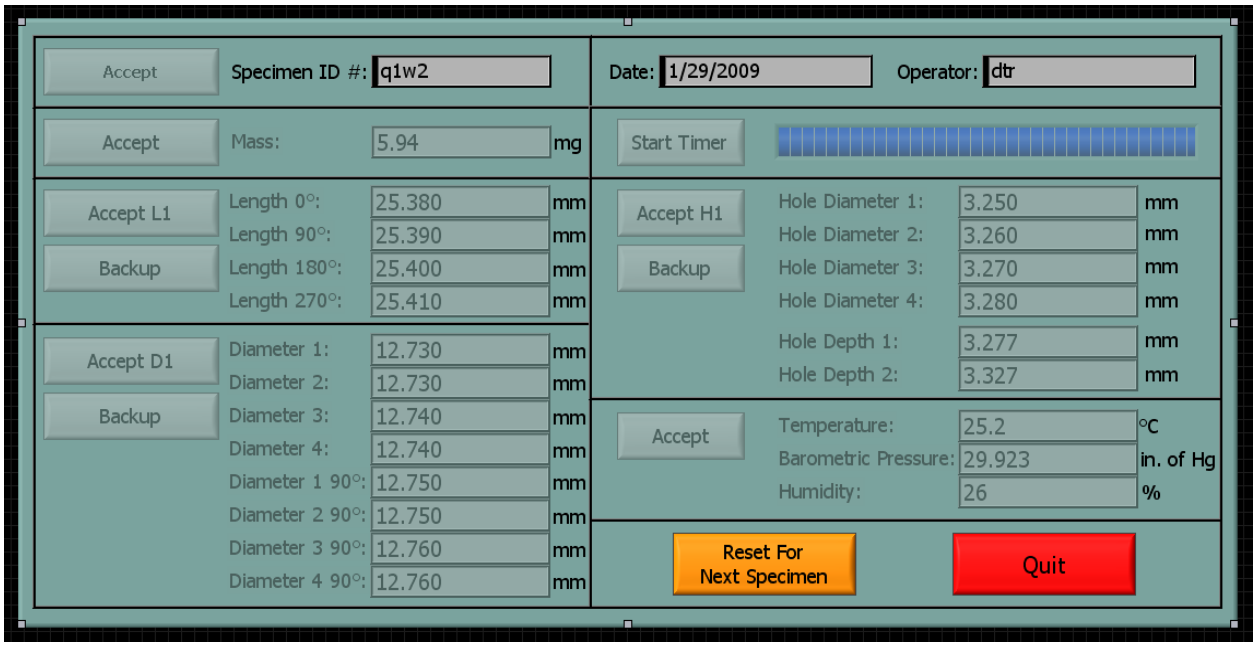


Figure 6. Physical and dimensional measurements program user interface.

3.1.1 Departures from ASTM C559-16

There are no departures from ASTM C559-16.

3.2 Electrical Resistivity

Electrical resistivity is used as a rapid, simple means to determine the grain orientation, structure, and crystallinity of graphite. In conjunction with optical microscopy, it can be used to determine the microstructural texture of graphite components with a minimum of specimen preparation work. Resistivity is measured following ASTM C611-98(2016), "Standard Test Method for Electrical Resistivity of Manufactured Carbon and Graphite Articles at Room Temperature." The measurement technique is commonly referred to as a 4-point probe. It consists of passing a known current through the specimen and measuring the voltage across the specimen at known locations.

Based on Ohms Law, the resistance is measured, and the resistivity is calculated from:

$$\rho = R \cdot A / L$$

where R is the measured resistance, A is the cross-sectional area, and L is the length over which the voltage is measured.

Figure 7 shows a test fixture fabricated at INL that allows a specimen to be rotated for multiple measurements of voltage around its periphery. The user interface for electrical resistivity is shown in Figure 8. The data acquisition screen steps the user through the acquisition process of the voltage and resistance measurements. The program communicates with both a Keithley Model 2182 nanovoltmeter and a Keithley Model 6220 DC precision current source. Using the current source, a known current is applied across the specimen, and the measured voltage is acquired by the software. During data collection, the application calculates the average voltage, average resistance, percent change of the resistance, and, ultimately, the resistivity. When all of the data has been taken, the results are recorded electronically in the applicable spreadsheet.

Uncertainty in the resistivity measurement is mainly comprised of the contact resistance between the specimen and the contacting blades for the voltage measurement. Specimen temperature and the temperature of other bi-metal junctions in the voltage measuring leads are also factors. These effects are minimized by passing the current through the specimen in two directions and averaging the measured voltage for each direction. In this way, any thermoelectric or small differences in junction resistances will cancel out. A round-robin test series reported in the ASTM C611-98(2016) precision and bias section states a lab-to-lab variability of 2.5%. A round-robin test series such as this would take into account the variables discussed above and is considered a good estimate of the measurement uncertainty.

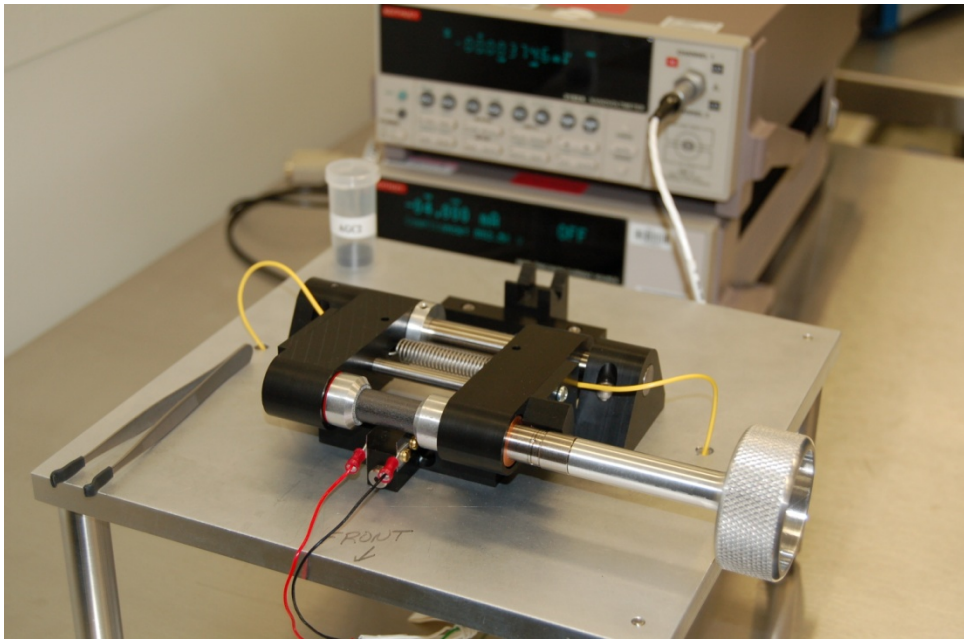


Figure 7. Electrical resistivity measurement station.

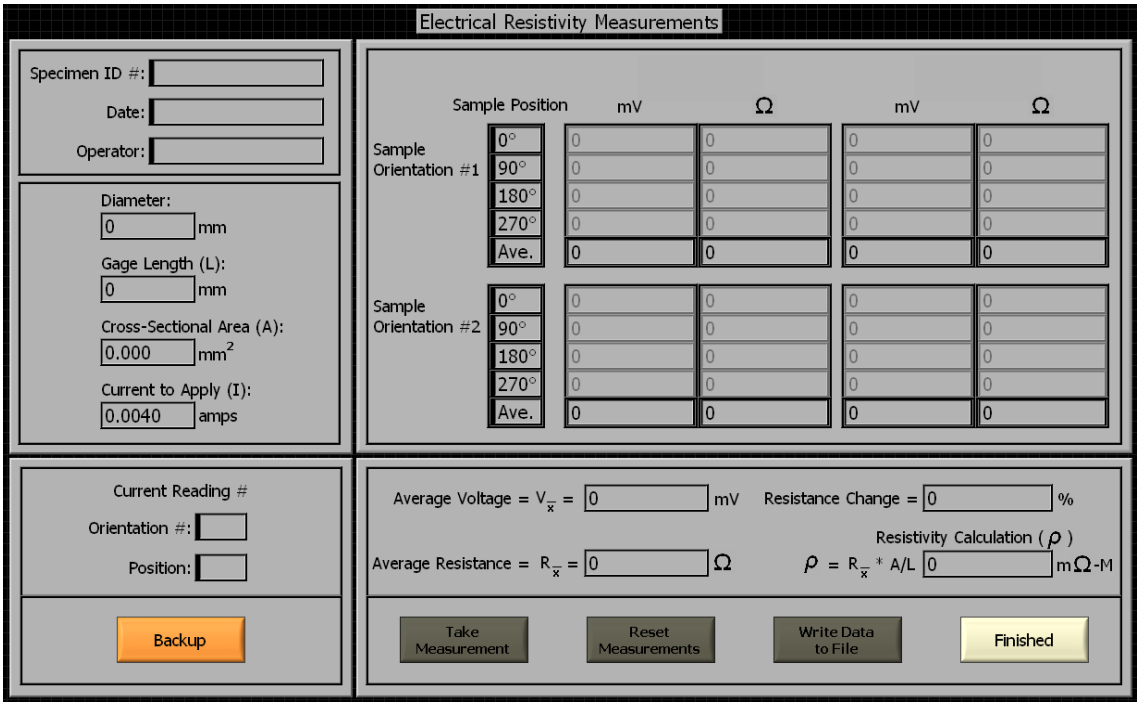


Figure 8. Software user interface for electrical resistivity.

3.2.1 Departures from ASTM C611-98(2016)

Departures from ASTM C611-98(2016) include:

- *Paragraph 5.2.3*. The small size of the AGC samples constrains the length-to-diameter ratio for resistivity specimens to 2:1, which is smaller than the ASTM recommended ratio of 6:1. This

difference is not expected to influence the measurement. Figure 7 above shows a specialized fixture designed for the geometry of the AGC specimens.

- *Paragraph 5.2.3.1*. The small size of the AGC samples constrains the gauge length-to-diameter ratio to 1:1, which is smaller than the ASTM recommended ratio of 4:1. The effect of this departure in the measurement is well within the measurement accuracy.
- *Paragraph 7.2*. The standard requires the heating effect on specimen resistivity to be less than 0.1%. Previously, the current would be continuously applied to the test specimen before, during, and after the measurement. The continuous application of current joule heating is a distinct possibility. However, the technique employed for the AGC specimens utilizes a pulsed current of 4 mA. In this way, the current is only applied long enough to take the voltage measurement, and the potential for joule heating is eliminated.

  To investigate heating, samples were tested by measuring the resistance eight times more than what would normally occur, giving ample opportunity for sample temperature changes to affect its resistance. The experiment gathers 64 resistance measurements on three different samples. Each of the three samples was tested eight times continuously, with each resistance measurement consisting of a separate forward current and reverse current measurement. Shown in Figure 9 is a representative plot of the data that was gathered from a single sample over the time period it took to perform the 64 measurements. The linear regression shows a slight increase in resistance. Using this regression for the eight measurements of a single specimen, one would expect an increase in measured resistance of less than 0.02% for a single specimen measurement.

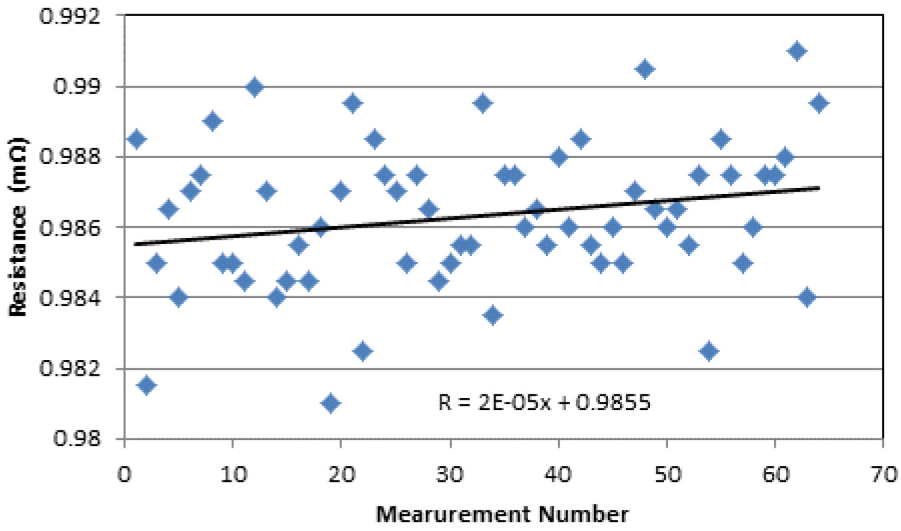


Figure 9. Calculated resistance from 64 current-voltage measurements on the same sample. This is equivalent to performing the complete ASTM procedure on the same sample eight times in a row.

3.3 Approximation of Elastic Modulus from the Measurement of Sonic Velocity

The mechanical properties of graphite are necessary to determine the structural integrity of graphite components. These properties are essential to determining the structural strength and integrity of the reactor core. The as-received and irradiated values are needed for whole-core models, which will be used for the graphite design code. This elastic modulus test is carried out in accordance with ASTM C769-98(2005), “Standard Test Method for Sonic Velocity in Manufactured Carbon and Graphite

Materials for Use in Obtaining an Approximate Value of Young's Modulus." In this measurement, the transmitting piezoelectric transducer sends a 2.25 MHz sound wave through the specimen. At the opposite end of the specimen, the acoustic wave is received by another piezoelectric transducer. The sonic velocity of the specimen is the ratio of specimen length to the signal time lapse between transducers.

An approximate value for Young's modulus, E, can be obtained from:

$$E = \rho V^2$$

Where ρ is the specimen density and V is the sonic velocity.

Figure 10 shows the sonic velocity measurement station. In the foreground are the fixtures for clamping the specimen between the transducer and receiver, which were specifically designed and fabricated at INL for this application. They have unique features that improve measurement accuracy, precision, and efficiency. Specimens are easily and rapidly loaded into the fixture using the cam-operated clamp. Measurement precision is improved because the spring-loaded clamp applies consistent pressure between the transducers and specimen, resulting in repeatable couplet thickness.

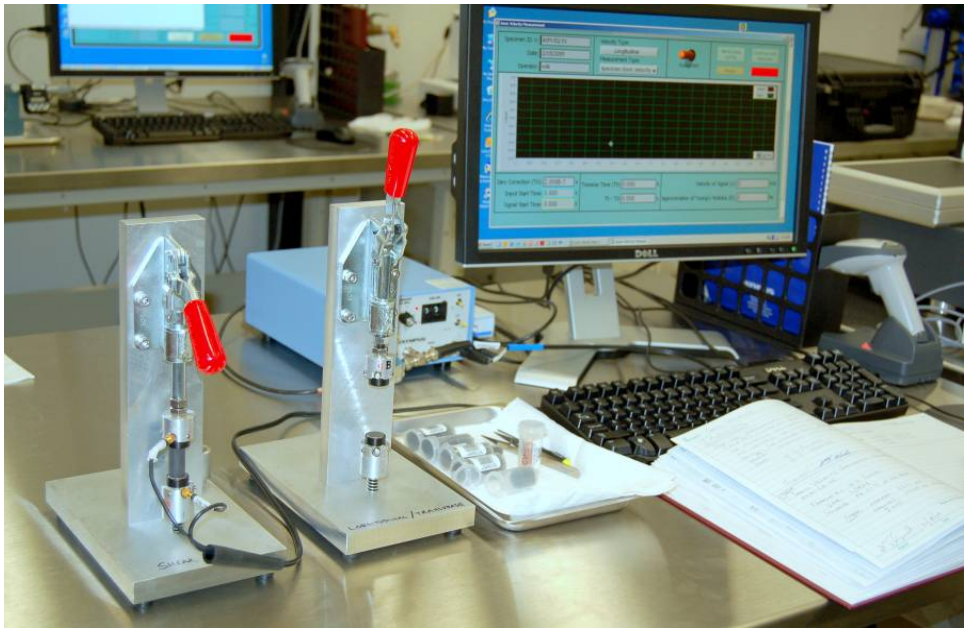


Figure 10. Sonic velocity measurement station.

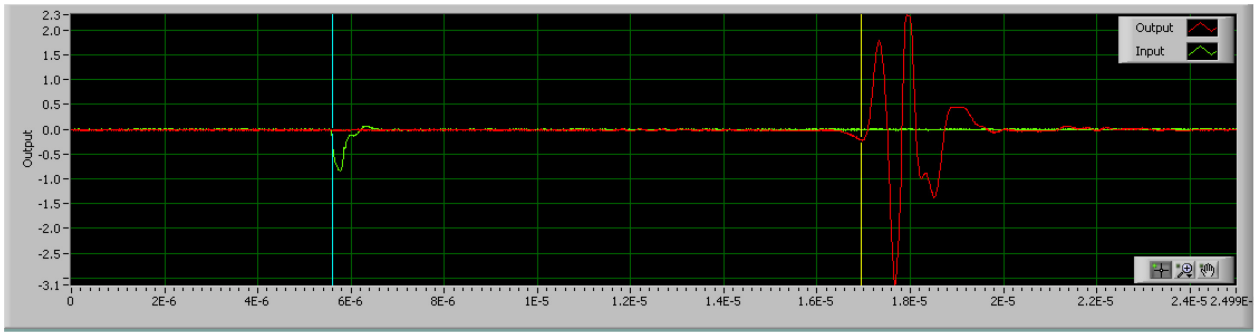
As specified in Paragraphs 8.1 and 8.5.1 of C769-98(2005), a suitable coupling medium should be used and reported with the data. Here, Shear Gel, manufactured by Sonotech Inc., is used for a shear wave couplet, and Ultragel II, also manufactured by Sonotech Inc., is used for the transverse wave couplet.

Figure 11 shows the LabVIEW software user interface display for sonic velocity measurements after scanning the barcode of the specimen to be tested. This screen is used to acquire sonic velocity measurements of a specimen in both the longitudinal and shear directions. Operating much like an oscilloscope, the cursors automatically mark the time between the transmitted wave and the received wave. Also shown in Figure 11 are two examples of the shear wave and transverse wave timing locations for properly coupled specimens. The specimen length divided by this transit time is the sonic velocity.

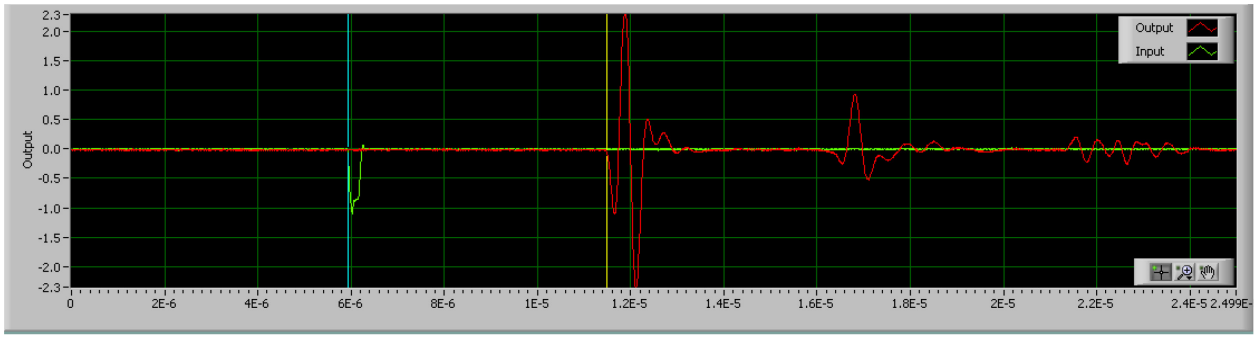
The uncertainty in determining elastic moduli from the measurement of sonic velocity comes from several sources. First there is the effect of material and geometry related dispersion of the transmitted wave. ASTM C769-98(2005) provides guidance to minimize this problem by assisting in the choice of

the correct frequency. This technique also assumes linear elastic behavior, but graphite is not a completely linearly elastic material. And finally, the operator's judgment on the placement of the timing cursors is somewhat subjective. Clean wave forms to base these judgments on are highly dependent on the quality of the transducer-material coupling.

These sources of error are difficult to quantify; therefore, they are difficult to combine in a propagation of error analysis. However, ASTM C769-98(2005) describes in some detail a round-robin test series between different laboratories. Using round-robin test data to determine a coefficient of variation (COV) provides a good means of estimating the measurement uncertainty. With caution, the COV of 3.8% reported in ASTM C769-98(2005) is taken here to be representative of the uncertainty of these measurements. When considering a single material and making comparisons between the pre- and post-irradiation values, the precision of these measurements is good enough to consider differences greater than 4% significant. However, one is cautioned to refrain from using the values here as absolute, or better than ± 10% accurate.



Shear wave timing.



Transverse wave timing.

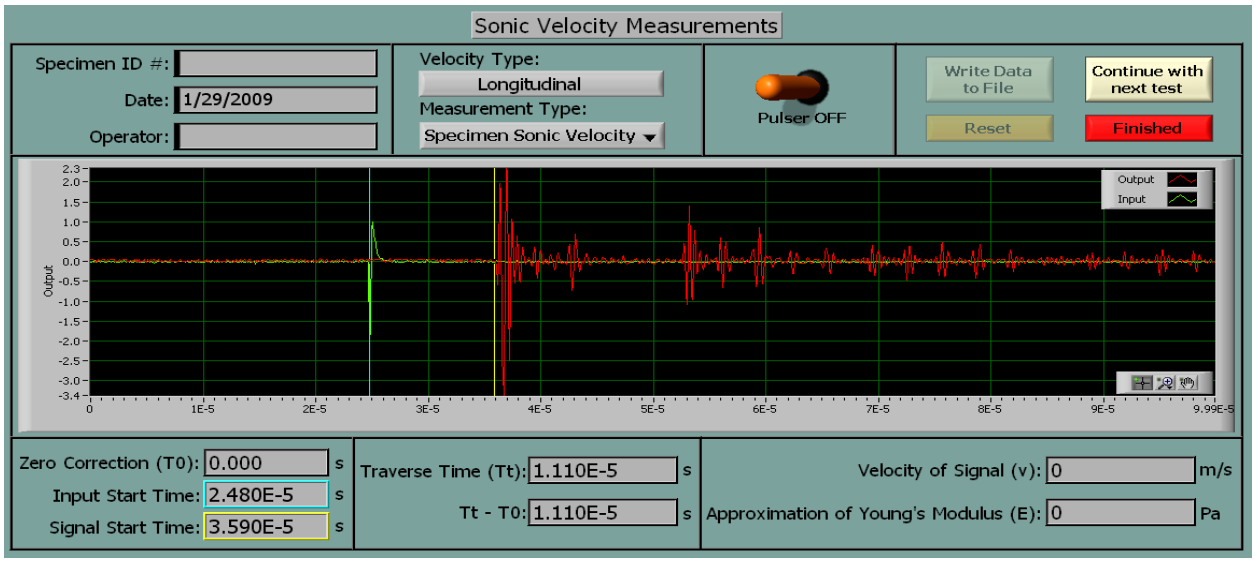


Figure 11. Sonic velocity measurement user interface.

3.3.1 Departures from ASTM C769-98(2005)

There are no departures from ASTM C769-98(2005).

3.4 Modulus of Elasticity by Fundamental Frequency Measurement

As previously discussed, the mechanical properties of graphite are necessary to determine the structural integrity of reactor core structure by assessing the strength and integrity of the graphitic components. This test method measures the fundamental resonant frequency of test specimens of suitable geometry by exciting them mechanically with a singular elastic strike. Specimen supports, impulse locations, and signal pick-up points are selected to induce and measure specific modes of the transient vibration of the specimen. The transient signals are analyzed, and the fundamental resonant frequency is isolated and measured by the signal analyzer. The measured fundamental resonant frequency, specimen dimensions, and mass are used to calculate the dynamic Young's modulus, shear modulus, and Poisson's ratio per ASTM C747-16, "Standard Test Method for Moduli of Elasticity and Fundamental Frequencies of Carbon and Graphite Materials by Sonic Resonance." The fundamental frequency measurement station is shown in Figure 12.

Figure 12. Fundamental frequency measurement station.

After the specimen's barcode has been scanned and the data acquisition program confirms that fundamental frequency data does not already exist for that specific specimen, the user interface for fundamental frequency measurements are displayed, as shown in Figure 13.

Figure 13. User interface for fundamental frequency measurements.

After placing a specimen in the test fixture, the user excites it by lightly tapping it with a small mechanical impulse. The specimen is supported in such a way that it vibrates at its natural frequency. A microphone placed underneath one end of the specimen in combination with the GrindoSonic frequency processing electronics measure this frequency, which is recorded and displayed by the computer. The modulus of elasticity is calculated and displayed next to the newly acquired frequency. If the results are satisfactory, the user can press the "Save 1st Frequency" button and go on to the next measurement. Following the recommendations of ASTM C747-16, ten readings of the fundamental frequency are measured and averaged before the results of the test are recorded.

ASTM C747-16 describes in detail a round-robin test series using graphite materials, along with an analysis of the propagation of errors in the calculation of moduli from the measurement of resonant frequency, geometry, and mass of the specimen. This error analysis shows the major sources of experimental variation are the measurement of the fundamental frequency and the smallest dimension (diameter) of the specimens due to their higher exponent in the modulus calculations. Both the propagation of error analysis and round-robin data indicated an uncertainty of less than 2% for this test method. However, the creep specimens tested here do not meet the geometry requirements of ASTM C747-16. With a length-to-diameter ratio of only 2, these specimens are, at times, difficult to excite consistently and in a single mode of vibration. Once a resonant frequency is determined by an experienced operator for the flexural mode of vibration, it was easily repeated within 2% uncertainty.

3.4.1 Departure from ASTM C747-16 include:

- *Paragraph 7.3*. Requires that the length-to-diameter ratio (L/d) be greater than 5. The L/d ratio for AGC creep specimens is 2, and the empirical constant (A_c) used in the calculation of the modulus cannot be determined. The manufacturer of the equipment being used in this measurement has stated that consistent and accurate values of fundamental frequency can be measured for L/d = 2. Personnel

performing the measurement can determine that a consistent fundamental frequency is obtained by viewing the displayed frequencies, shown in Figure 13.

3.5 Thermal Expansion

Understanding the thermal expansion behavior of graphite components, represented as the CTE, is critical for determining dimensional changes that occur as a result of changing temperatures and temperature gradients. Localized internal and external stresses can be imposed on graphite core components as they experience differential thermal expansions. The irradiation dimensional change response is also affected by the thermal expansion, with grades exhibiting a higher CTE that have lower turnaround neutron dose levels. Finally, the thermal expansion is highly dependent upon the graphite microstructure, such as orientation/anisotropy, pore size and distribution, and crystallinity. Determining the extent of the CTE change as a function of irradiation dose and temperature will be a key parameter for the reliable calculation of stress states within graphite components, volumetric changes, and irradiation creep rates.

CTE is measured from ASTM standard E228-17. This test method uses a push-rod-type dilatometer to determine the change in length of a graphite specimen relative to that of the specimen holder as a function of temperature. The temperature is varied over the desired range at a slow constant heating or cooling rate. The mean coefficient of thermal expansion, α, is calculated from the measured data using:

$$\alpha = \frac{1}{L_0}\frac{\Delta \mathrm{L}}{\Delta \mathrm{T}}$$

Where L_0 is the specimen initial length, ΔL is the change in length, ΔT is the temperature difference between a specified reference temperature and the temperature at which the change in length was measured.

The Netzsch DIL 402 C commercial system (Figure 14) currently used at INL does not have the capability to cool the specimen below the ambient temperature. Therefore, the initial length at 20°C is linearly extrapolated from expansion data between 100°C and 150°C, and the mean CTE is calculated from a 20°C reference temperature.

It is important to note that the CTE of irradiated specimens can only be measured up to their irradiation temperature. Taking the specimen above the temperature at which it was irradiated would most likely result in some annealing of the irradiation damage and thus change its physical properties. For AGC-4, the average nominal irradiation temperature is 800°C ± 50°C; therefore, the maximum temperature for initial CTE measurements will be 700°C.

The greatest source of experimental error in the dilatometry test method described here is the correction made for the expansion of the specimen holder and push rod/LVDT mechanism. This differential between the specimen and the apparatus must be considered to isolate only the specimen expansion. Studies reported in the precision and bias section of ASTM E228-17 have indicated that this type of dilatometry can be accurate to 4% when calibrations are performed carefully.

Figure 14. Commercial push rod dilatometer for measurement of CTE.

3.5.1 Departure from ASTM E228-17

A departure from ASTM E228-17 is presented in Paragraph 7.1.2.3. The Netzsch Dialometry 402°C commercial system currently used at INL does not have the capability to cool the specimen below the ambient temperature. Therefore, the initial length at 20°C is linearly extrapolated from expansion data between 100 and 150°C, and the mean CTE is calculated from a 20°C reference temperature.

3.6 Thermal Diffusivity

The ability to conduct heat through the graphite core is critical to the operational efficiency of the HTR and for the passive removal of decay heat during cooldown. Reduction of the thermal conductivity within graphite can have a significant effect on the passive heat removal rate and thus the peak temperature that the core and, subsequently, the fuel particles will experience during off-normal events. Determining changes to the conductivity as a function of irradiation dose and temperature is important for the safety analysis. Here, ASTM E1461-13 is followed for the calculation of thermal diffusivity and conductivity. Thermal diffusivity (δ) is measured and defined as the ratio of thermal conductivity to volumetric heat capacity by:

$$\delta = \frac{k}{\rho C_P}$$

where k is the thermal conductivity, ρ is the density, and C_P is the specific heat.

Diffusivity measurements are performed on the button shaped piggyback specimens. A pulsed laser is used to subject one surface of the specimen to a high-intensity, short-duration energy pulse. The energy of this pulse is absorbed on the front surface of the specimen and the resulting rise in rear-face temperature is recorded. The thermal diffusivity is calculated from the specimen thickness and the time required for the rear-face temperature to reach 50% of its maximum value. A commercially available laser flash apparatus (LFA), complete with vendor-developed software for instrument control and data acquisition, is used as shown in Figure 15.

Uncertainty in the measurement of thermal diffusivity arises from specimen heat loss and the temperature measurement. Specimen temperature measurement is performed with a calibrated type-S

thermocouple in the near vicinity of the specimen. Being relatively straight forward, the specimen temperature measurement is typically only a small contribution to the overall measurement error or uncertainty.

The primary contributor to the measurement uncertainty is heat loss from the specimen. Because this measurement technique depends on the assumption of a one-dimensional heat transfer from the flat face receiving the laser pulse to the flat face radiating to the detector, heat loss errors are mainly attributed to radiative heat loss from the circumference of the specimen at temperatures above 300°C. Typically, several correction models are provided with the instrument software to account for this heat loss. As the specimen diameter to thickness ratio decreases, the heat loss increases to the point that the correction models can no longer account for the error. A study was performed to gain a fuller understanding of the limits of the models made available with the NETZSCH LFA and the dependence of the diameter to thickness ratio on measurement error. In this study, the heat loss models were applied to data taken on specimens with various diameter-to-thickness ratios and at specimen temperatures between 25°C and 1000°C. It was determined that the Cowan model, along with diameter to thickness ratios greater than or equal to 2, resulted in the determination of the diffusivity within ASTM E1461-13 and the manufactures specified uncertainties of 4% and 3%, respectively. This was further verified by instrument functional tests performed monthly on a pure iron validation specimen for which the diffusivity was determined to be within 3% of the Touloukian values between 100°C and 700°C.

As with determining the CTE, the diffusivity of irradiated specimens can only be measured up to their irradiation temperature. Taking the specimen above the temperature at which it was irradiated would most likely result in some annealing of the irradiation damage and thus change its physical properties. For AGC-4, the average nominal irradiation temperature was 800°C ± 50°C; therefore, the maximum temperature for initial measurements will be 700°C.

Figure 15. Laser flash apparatus measurement station for determination of thermal diffusivity.

3.6.1 Departure from ASTM E1461-13

A departure from ASTM E1461-13, presented in Paragraphs 11.1 and 12.1.5, is that the Netzsch Instrument software only calculates thermal diffusivity at the 50% rise time. This has no impact on the resulting diffusivity value and is commonly accepted as the only time needed to accurately calculate the diffusivity.

4. REFERENCES

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