Proceedings of the ASME 2023
Pressure Vessels & Piping Conference
PVP2023
July 16-21, 2023, Atlanta, Georgia, USA

PVP2023-106610

DEVELOPMENT OF A GRAPHITE IRRADIATION QUALIFICATION PLAN FOR THE XE-100 REACTOR*

Anne A. Campbell[1], Josina W. Geringer[1], Adrian M. Schrell[1], Samuel Baylis[2], Timothy Lucas[2], Peter J. Pappano[3], Martin van Staden[2]

[1]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN
[2]X Energy, LLC., Rockville, MD
[3]TRISO-X, LLC., Rockville, MD

ABSTRACT

The American Society of Mechanical Engineers Boiler and Pressure Vessel Code, Section III "Rules for Construction of Nuclear Facility Components", Division 5 "High Temperature Reactors", Subsection HH "Class SN Nonmetallic Core Components", Subpart A "Graphite Materials", has a list of materials properties that must be measured and included in the materials data sheet (MDS) for each grade of graphite to be used in a reactor core. Two sections of the MDS includes measuring the effects of degradation, due to oxidation or neutron irradiation damage, on these materials properties. A program at Oak Ridge National Laboratory (ORNL), supported by X Energy, LLC., began in 2017 to produce the irradiation-induced property change results of graphite grades that will be used in the X Energy Xe-100 small modular reactor. Significant effort has been performed around the world to capture these property changes for a range of graphite grades through both the US Advanced Graphite Creep program, and the EU INNOGRAPH/ARCHER program. But both programs were primarily targeted towards higher temperature reactor designs that were of greater importance at the beginning of both programs resulting in a lack of data for temperatures below 600°C. The program at ORNL has been developed to produce the data necessary to assist with completing the irradiation effects on materials properties of the MDS for the graphite grades of interest for X Energy. This paper will discuss the design of the irradiation program including the irradiation conditions (neutron fluence and irradiation temperatures), the types of specimens being used in this program, and the irradiation methodology.

Keywords: Nuclear graphite, ASME Code Qualification, Irradiation effects

1. INTRODUCTION

X Energy, LLC., is designing a small modular nuclear reactor called the Xe-100. This reactor design is based on the gas-cooled high temperature reactor (HTR) thermal reactor concept within the Gen-IV reactor designs. The reactor uses TRISO particle fuel formed into pebbles, helium as the coolant, and graphite as both the core structural material and neutron moderator. Each reactor core is designed with a 200 MW thermal and 80 MW electric output.

The use of graphite as the core structural support results in the use of graphite within the purview of American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code, Section III "Rules for Construction of Nuclear Facility Components", Division 5 "High Temperature Reactors" (BPV III-5) [1]. Subsection HH, Subpart A discusses the requirements for Graphite materials, specifically HHA-III-3000 discusses the properties to be measured on as-manufactured and degraded graphite (degradation due to oxidation or irradiation damage) while Article HHA-II-2000 provides the materials data sheet (MDS) to be filled in with these properties.

X Energy approached researchers at Oak Ridge National Laboratory (ORNL) to collaborate and aid in the development and execution of a research program that would produce the data necessary to allow X Energy to complete the MDS for the graphite grade selected for the Xe-100 core. This paper will discuss the irradiation program that has been developed in support of this effort.

2. ASME BPV-III-5 REQUIREMENTS

The requirements for quantifying the changes to graphite material properties due to irradiation are listed in Article HHA-III-3300. The required changes to materials properties include dimension, elastic modulus, strength, thermal conductivity, coefficient of thermal expansion, creep coefficient, and stored energy but only when a portion of the graphite will experience temperatures at or below 200°C. The changes in these properties must be captured at a range of neutron doses and irradiation temperatures that encompass the temperature and dose combinations that the graphite will experience during the reactor lifetime. Performing an irradiation program to adequately quantify these irradiation-induced property changes can result in programs costing more than $15M and lasting longer than 10 years [2].

3. IRRADIATION PROGRAM DEVELOPMENT

The first step for developing an irradiation program to complete the MDS is to understand the expected temperature and neutron fluence conditions the graphite will experience over the reactor lifetime. A hypothetical temperature and dose range for the Xe-100 reactor was provided by X Energy, which ORNL expanded to be fully captured in the irradiation program envelope (the blue field is the expanded envelope in Figure 1).

The second step was to determine if any previous or on-going research could be used to supplement the data needs for the graphite grade selected by X Energy. The INNOGRAPH [3] program performed at the High Flux Reactor (HFR) in Petten published results on graphite irradiated at temperatures of 750°C and 950°C and maximum doses of 22 displacements per atom (dpa) at 750°C and 13 dpa at 950°C. The U.S. DOE sponsored Advanced Graphite Creep (AGC) and High Dose Graphite (HDG) irradiation program [4] is currently in progress in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) are producing results for 600°C and 800°C with a maximum dose of 15 dpa for both temperatures. The conditions encompassed by these two programs are highlighted in Figure 1. Unfortunately, both INNOGRAPH and the AGC/HDG programs were both started when the primary focus for gas-cooled reactors was on very high temperature systems, as such most of the data from these programs are not directly relevant for the needs related to the Xe-100.

The final step is to determine the irradiation temperature and fluence conditions to target for this program (red circles in Figure 1). These irradiation conditions are based on temperature and fluence bounds provided by X Energy. Article HHA-II-4000 requires that data be sufficiently dense to provide confidence for interpolation of the results, and Article HHA-III-3300 states a maximum of 200°C between irradiation temperatures. At 200°C the behavior of graphite exposed to neutron irradiation is fundamentally different than at temperatures at or above 300°C, so for the 200°C temperature five fluence steps were selected. For temperatures of 300°C to 600°C three temperatures have been targeted, with a 150°C difference between temperatures. For these higher temperatures the desired fluences are spaced at roughly 5 dpa intervals providing six fluence points for 300°C and 450°C and 4 dpa for the four fluences for 600°C.

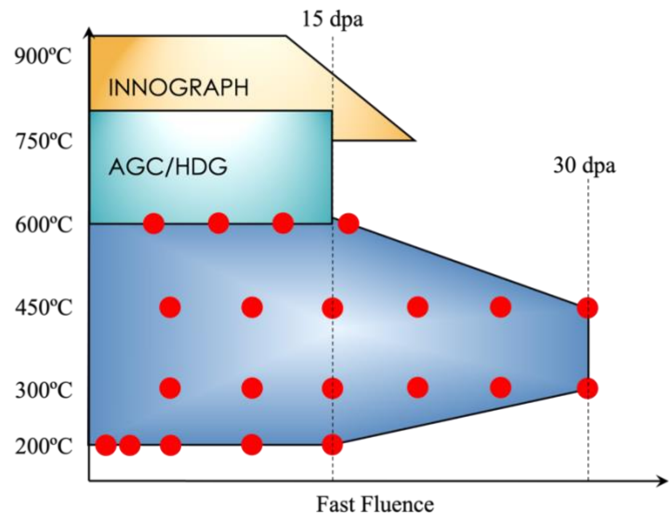

FIGURE 1: FLUENCE AND TEMPERATURE RANGE ENVELOPE PROPOSED BY ORNL FOR THE XE-100 REACTOR (BLUE FIELD), DISCRETE TEMPERATURE AND FLUENCE CONDITIONS FOR ORNL IRRADIATION PROGRAM (RED DOTS), AND IRRADIATION CONDITIONS COVERED BY INNOGRAPH [3] AND AGC/HGD [4] IRRADIATION PROGRAMS.

3.1 Irradiations

The irradiation of graphite in this program will occur in the flux trap of the ORNL High Flux Isotope Reactor (HFIR). The HFIR is operated at a constant power of 85 MW with a typical operating cycle length of 24-25 days. Graphite irradiated in the HFIR flux trap can accumulate nominally 1.2-1.4 dpa per reactor cycle.

The graphite will be irradiated in small capsules that fit within the HFIR flux trap. These capsules are roughly 60 mm long and 11 mm in diameter, so the space within a capsule for specimens is roughly 6×6×48 mm. These small capsules are not able to be instrumented, and instead the temperature of the specimens is controlled via passive methods. Passive temperature control is achieved by tailoring the width of a gas gap between the inner pieces of the capsule (specimens and specimen holder) and the outer capsule housing that is in contact with the HFIR coolant, and the fill gas used. The primary assumption for these passively controlled capsules is that the constant operating power of the HFIR results in a nearly constant gamma flux within the flux trap positions during a cycle and from cycle to cycle.

The capsules are modeled in the ANSYS three-dimensional finite element analysis (FEA) software. During modeling the capsules are optimized for the necessary gas gap and inert fill gas to achieve the desired specimen temperatures, which also accounts for the different gamma fluxes/heating rates at different positions within the flux trap. Determination of the actual irradiation temperature is performed via analysis of the annealing behavior of silicon carbide (SiC) pieces that are

included in the irradiation capsules as passive temperature monitors. The annealing of the irradiation damage in the SiC is done in via a dilatometry measurement method developed at ORNL [5], or a similar method that utilizes an isochronal annealing method developed at ORNL [6]. Once the irradiation temperature of the SiC is known, the temperature of the graphite specimens are determined by comparing the modeled and measured SiC temperatures with the graphite temperatures from the FEA models of the capsule.

3.2 Specimen Geometries

The available space for specimens within the irradiation capsules is the primary limiter for the geometries and size selection of irradiation specimens. Additionally, being able to measure multiple properties from a single specimen is preferred to keep the total number of specimens requiring neutron irradiation as low as feasible. Three specimen geometries are being used for this irradiation program. All specimens have a diameter of 6 mm and thicknesses of 1 mm, 3 mm, and 12 mm.

3.3 Specimen Size Effect

In an ideal scenario, the irradiation specimen sizes would be sufficiently large as to conform with the minimum specimen dimensions listed in the various ASTM test standards. But as occurs in most irradiations in materials test reactors, using specimens that conform to ASTM sizes is not possible in this program. To account for this, a study is being performed in accordance with ASTM D7775 [7] to understand how the properties measured on the reduced-size specimens relate to the values when measured on specimens that conform to standard requirements.

The Ø6×1 mm specimens are only used in the 200°C irradiation because at this temperature the primary property to measure is the stored energy buildup with the graphite and this is the optimal specimen size for the differential scanning calorimeter that will be used to measure the stored energy [8]. There is no expected size effect for stored energy measurements so there is no experimental effort to quantify this.

The Ø6×3 mm specimens will be utilized to quantify the changes in dimensions [9], thermal conductivity (measure thermal diffusivity [10] and calculate conductivity via annex A.6 in [11]), and strength via a disk compression method [12]. The effect of specimen size on the properties for these disks are being quantified for these properties with specimens as large as Ø30×15 mm and as small as Ø5×2.5 mm.

The Ø6×12 mm specimens will be utilized to quantify the changes in dimensions [9], elastic properties [13], coefficient of thermal expansion [14], and compression strength [15]. The effect of specimen size on the properties for these rods are being quantified for these properties with specimens as large as Ø24×48 mm and as small as Ø5×10 mm.

3.4 Capsule Design/Configuration

Two different capsules designs are being utilized in this effort (Figure 2). The first design is specifically configured for the “Low Temperature” 200°C irradiation temperature (top image in Figure 2) contains a specimen stack consisting of ten Ø6×1 mm disks and eight Ø6×3 mm disks. The second “Multi-Use” capsule is configured for the three other irradiation temperatures (bottom image in Figure 2) contains six Ø6×3 mm disks and two Ø6×12 mm rods.

One primary question related to these small specimens is whether properties measured are representative of the bulk material. This is being addressed by the fact that for the 200°C capsules there are ten Ø6×1 specimens for stored energy measurement and four of the Ø6×3 for each orientation. This gives ten specimens for stored energy measurements and four replicate specimens for thermal conductivity measurements for each orientation.

For the higher irradiation temperatures, there are three identical capsules being irradiated for each temperature/fluence combination. This will result in nine Ø6×3 specimens for each orientation and three Ø6×12 rods for each orientation. This means that for strength measurements, which are more prone to a larger statistical spread, there will be a larger statistical body of data for analysis and to account for these extremely small specimens relative to the bulk sizes of graphite billets.

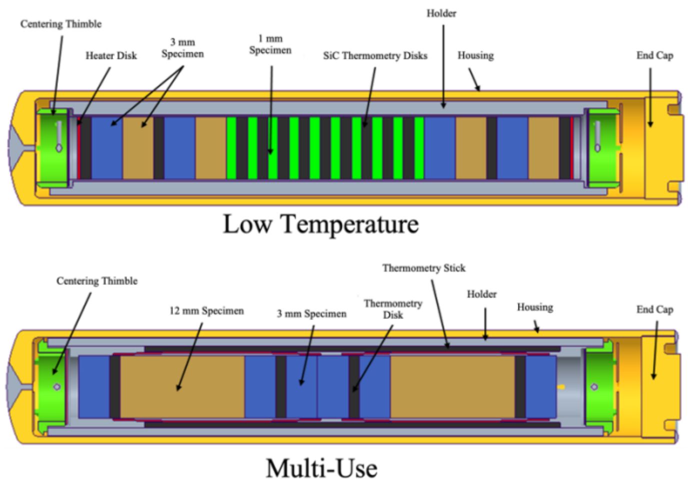

FIGURE 2: CAPSULE DESIGNS FOR LOW TEMPERATURE IRRADIATION (TOP) AND MULTI-USE FOR 300°C-600°C (BOTTOM).

The capsules for this current irradiation program do not include the capability to apply a stress to the samples to capture irradiation creep at these temperatures. Currently the only program performing irradiation creep is the AGC/HDG program at INL [4]. It is understood that there will be a need to perform irradiation creep experiments within the temperature and fluence envelope of this program, but the extent of a creep program is still under discussion.

4. SUMMARY

An irradiation program is underway at ORNL that will produce data that will be used to assist in completion of the MDS for graphite grades that will be used in the Xe-100 small modular reactor. Specimens are being irradiated, without stress, in

capsules designed for the ORNL HFIR flux trap. The irradiation temperatures range from 200°C to 600°C with dose ranging from 1 to 30 dpa. Three specimen geometries are being utilized to measure the changes to materials properties including dimensions, elastic properties, strength, thermal conductivity, coefficient of thermal expansion, and stored energy.

ACKNOWLEDGEMENTS

Support for this work was provided by the U.S. Department of Energy, Office of Nuclear Energy via the X-energy Advanced Reactor Concepts (ARC) 15 Award. Oak Ridge National Laboratory is managed by UT-Battelle, LLC, under contract No. DE-AC05-00OR22725 for the U.S. Department of Energy.

REFERENCES

[1] "ASME Boiler and Pressure Vessel Code An International Code, SECTION III Rules for Construction of Nuclear Facility Components, Division 5 High Temperature Reactors", ASME, New York, NY,

[2] Campbell, A.A., "Perspective on “code qualifying” new graphite grades for use in advanced nuclear reactors*", *Frontiers in Nuclear Engineering*, **1**, (2022) 1045607.

[3] Heijna, M.C.R., de Groot, S., and Vreeling, J.A., "Comparison of irradiation behaviour of HTR graphite grades", *Journal of Nuclear Materials*, **492**, (2017) 148-156.

[4] Windes, W., "ART Advanced Graphite Creep (AGC) Irradiation Experiment", Presented at *DOE-NE Advanced Reactor Technologies 2022 Gas-Cooled Reactor Program Review Meeting*, Virtual, July 12-14, 2022.

[5] Campbell, A.A., Porter, W.D., Katoh, Y., and Snead, L.L., "Method for Analyzing Passive SiC Thermometry with a Continuous Dilatometer to Determine Irradiation Temperature", *Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms*, **370**, (2016) 49-58.

[6] Wang, H., Koyanagi, T., Geringer, J.W., Campbell, A.A., and Katoh, Y., "Determination of neutron irradiation temperatures of SiC using electrical resistivity method", *Journal of Nuclear Materials*, **540**, (2020) 152370.

[7] ASTM D7775-21, 2021, "Standard Guide for Measurements on Small Graphite Specimens", ASTM International, West Conshohocken, PA, DOI: 10.1520/D7775-21, www.astm.org.

[8] ASTM E1269 − 11 (2018), 2018, "Standard Test Method for Determining Specific Heat Capacity by Differential Scanning Calorimetry", ASTM International, West Conshohocken, PA, DOI: 10.1520/E1269-11R18, www.astm.org.

[9] ASTM C559-16 (2020), 2020, "Standard Test Method for Bulk Density by Physical Measurements of Manufactured Carbon and Graphite Articles", ASTM International, West Conshohocken, PA, DOI: 10.1520/C0559-16R20, www.astm.org.

[10] ASTM E1461-13 (2022), 2022, "Standard Test Method for Thermal Diffusivity by the Flash Method", ASTM International, West Conshohocken, PA, DOI: 10.1520/E1461-13R22, www.astm.org.

[11] ASTM C781-20, 2020, "Standard Practice for Testing Graphite and Boronated Graphite Materials for High-Temperature Gas-Cooled Nuclear Reactor Components", ASTM International, West Conshohocken, PA, DOI: 10.1520/C0781-20, www.astm.org.

[12] ASTM D8289-20, 2020, "Standard Test Method for Tensile Strength Estimate by Disc Compression of Manufactured Graphite", ASTM International, West Conshohocken, PA, DOI: 10.1520/D8289-20, www.astm.org.

[13] ASTM C769-15 (Reapproved 2020), 2020, "Standard Test Method for Sonic Velocity in Manufactured Carbon and Graphite Materials for Use in Obtaining Young’s Modulus", ASTM International, West Conshohocken, PA, DOI: 10.1520/C0769-15R20E01, www.astm.org.

[14] ASTM E228-17, 2017, "Standard Test Method for Linear Thermal Expansion of Solid Materials With a Push-Rod Dilatometer", ASTM International, West Conshohocken, PA, DOI: 10.1520/E0228-17, www.astm.org.

[15] ASTM C695-21, 2021, "Standard Test Method for Compressive Strength of Carbon and Graphite", ASTM International, West Conshohocken, PA, DOI: 10.1520/C0695-21, www.astm.org.