ORNL/LTR-2024/3274

Update on Activities Related to the Library of Graphite Microstructures

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May 2024

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Materials Science and Technology Division

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Prepared by
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managed by
UT-BATTELLE LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

ABSTRACT

This report provides an overview and update of the ongoing efforts to create a comprehensive library of microstructures for nuclear graphite and carbon-based materials under consideration for nuclear applications. The library includes data on microstructural characterization of unirradiated graphite materials, a guide to the techniques used to analyze graphite (which complements the ASME guidelines and ASTM standards), a summary of characterization data for neutron-irradiated or oxidized material, and a compendium of microstructural information for carbon-based materials. These efforts are being conducted at various length scales for the filler and binder phases in graphite to better understand graphite's local structure and property relationships. This report is a follow-up to the previous milestone report titled *Report on initial development of a database of nuclear graphite characteristics based on microstructural characterization*, ORNL/TM/-2023/2992, published in July 2023.

The effort to develop the library of microstructures supports the US Department of Energy Office of Advanced Reactor Technologies program objectives of aiding the material selection, licensing, management, and core assessments of a graphite core by documenting the unirradiated microstructure of relevant grades or characterizing the microstructure's evolution under the reactor environment. Additionally, this project aims to provide (1) information and guidelines for the characterizing of graphite and (2) a protocol to assess a nuclear graphite grade.

1. INTRODUCTION

Graphite plays a critical role as a moderator and structural material for Generation IV advanced reactors, including the Very High-Temperature Reactor and various molten salt reactor designs. It is a complex material formed by filler and binder phases, and its properties depend on the characteristics and distribution of these phases. To confidently predict and inspect the long-term performance of graphite components in gas-cooled or molten salt environments, a deeper understanding of the fundamental relationships between processing, structure, and properties of graphites is essential. This understanding also includes knowledge about the availability, sourcing, and performance variations of candidate graphite grades.

The materials that can ensure the long-term reliability of this type of technology must be assessed and categorized to support the deployment of test reactors and the commercialization of novel nuclear power stations through graphite-moderated designs in the United States. The Oak Ridge National Laboratory (ORNL) Nuclear Graphite Team is creating a library of microstructure that characterizes and identifies the key phases, features, techniques, and materials for advanced reactors. This report is a continuation of the previous report [1] and publications that support the library of microstructures [2-6].

This report provides an update to four sections that comprise this research: (a) the library of microstructures of candidate graphite grades, (b) a library of defects caused by oxidation and/or neutron irradiation, (c) the guidelines to understand the different techniques commonly used to study graphite, and (d) a brief library of carbon materials used in the nuclear industry. These efforts will inform the industry about the characteristics of nuclear graphite and other carbon-based materials, provide additional guidance on the characterization of graphite that is not covered in standards or codes, inform surveillance campaigns of reactor cores, and enable the modeling and validation of stress analysis and fracture mechanics models.

Articles and reports are covered in the following subsections.

1.1 LIBRARY OF MICROSTRUCTURES OF CANDIDATE GRAPHITE GRADES

The selection of graphite materials and the design of components depends on the application and conditions generated by the reactor design. The microstructure characterization of nuclear grades provides valuable information about how different aging processes can degrade graphite. This portion of the project documents the microstructure of various graphites. The data generated in this section will help inform the quality control of graphite material and ensure consistency between batches of grades and support any assessments on the adequacy of the graphite materials.

The following articles and reports will support this section of the project:

1. "Serial sectioning optical microscopy of candidate graphite grades," journal article, status: in progress. This research will use serial mechanical polishing and optical microscopy to generate micrographs that characterize the microstructure of selected graphite grades. These images will help to understand the morphology of candidate graphite grades.

2. "Microstructural data of PCEA nuclear graphite," journal article, status: in progress. This journal publication will gather the microstructural data of three billets of PCEA nuclear graphite. Moreover, the data can be used to compare the differences in the graphite produced during three different batches.

1.2 LIBRARY OF DEFECTS CAUSED BY OXIDATION OR NEUTRON IRRADIATION

Although design and modeling establish multiple safety safeguards, the damage tolerance and safety margins for a reactor core and its respective graphite material must be understood. These limits might be directly assessed from reactor monitoring inspections. However, the degradation mechanisms are specific for each graphite material. This part of the project characterizes some of the main US candidate grades after they are degraded by oxidation or irradiation. This information can be used to establish boundaries for safe operation. Part of this characterization has been focused on the characterization of oxidation and neutron irradiation effects in nuclear graphite. Nuclear graphite components are subjected to cooling and thermal shock during refueling operations, and neutron irradiation induces defects and oxidation. This research will mimic some of these conditions to observe how the microstructure evolves as a result of degradation mechanisms. Moreover, this project investigates some features in graphite that can be used as forensic fingerprints and reveal the history of dimensional change in a graphite component. Materials Test Reactor irradiation campaign experiments are normally used as input information for material properties into design calculations. However, the microstructural information of these samples is not normally recorded. Linking the microstructural evolution with mechanical properties will enable better core behavior predictions.

The following articles and reports will support this section of the project:

1. "Microstructural characterization of the oxidation of nuclear graphite under chronic and accident conditions via conventional XCT and synchrotron XCT, journal article, status: in progress. This research produced a novel and systematic characterization of nuclear graphite's microstructural evolution under accident conditions or uniform oxidation. The findings elucidate how microstructure and pore connectivity influence the oxidation rate and microstructural evolution under these two types of oxidation conditions.

2. "FIB-SEM tomography of quinoline insoluble particles: a fingerprint of neutron irradiation effects of nuclear graphite", journal article, status: in progress. This research is conducting focused ion beam scanning electron microscopy tomography on unirradiated and neutron-

irradiated quinoline-insoluble particles to understand the densification of this graphite phase. Characterizing this phase of materials can be used as a forensic fingerprint of the irradiation effects in nuclear graphite.

1.3 GUIDELINES TO UNDERSTAND THE DIFFERENT TECHNIQUES COMMONLY USED TO STUDY GRAPHITE

Microstructural characterization is a key component of understanding neutron irradiation effects and graphite's performance. Several characterization techniques used to characterize graphite do not have clear guidelines for conducting the measurements. This part of the project is being conducted to support graphite microstructural research.

Articles and reports that will support this section of the project:

1. "Characterization of neutron irradiation effects, machining, polishing and oxidation in AXF-5Q graphite via Raman spectroscopy," journal article, status: in progress. Raman spectroscopy offers the potential to characterize diverse types of damage mechanisms in nuclear graphite and other carbon materials. This research aims to explore the sensitivity of Raman spectroscopy to various surface types and common damage mechanisms affecting nuclear. Specifically, the study compares the effect of irradiation temperature, neutron irradiation, and oxidation on polished, machined, and fractured surfaces of AXF-5Q graphite. By examining these effects on AXF-5Q graphite, a single-phase binderless graphite, the research aims to discern the influence of surface characteristics and irradiation conditions on Raman spectra.

2. "Benchmarking of microstructural characterisation methods for the characterisation of nuclear graphite, matrix graphite and pyrolytic carbon materials - Review of Laser Raman Spectroscopy", Organisation for Economic Co-operation and Development report, status: in progress. This report provides a general guideline of Raman spectroscopy's most important parameters and applications and their respective influence on graphite characterization.

3. "Intragrade Variations of Microstructural Characteristics and Distribution of Properties for Nuclear Graphites a case study for PCEA," journal article, status: in progress. Nuclear graphite's homogeneity and microstructural integrity can be influenced by both manufacturing techniques and the raw materials employed to produce the final product. Batches of graphite bricks produced by the same manufacturer commonly exhibit variations, even when the same manufacturer produces the same grade. Establishing protocols to assess the variations among batches of graphite is essential for qualifying this type of material and reusing legacy data. This research focuses on comparing three batches of PCEA graphite. Although the batches exhibit similar microstructural features, distinct differences may be attributed to variations in manufacturing processes during graphite production. The comparisons made in this study underscore the importance of identifying microstructural characteristics that most significantly affect the performance of graphite components in nuclear applications. This study investigates the variations in nuclear-grade graphite samples across different batches and proposes protocols to assess the significance of these differences. Techniques utilized include scanning electron microscopy, helium pycnometry, x-ray diffraction, x-ray computed tomography, and ultrasonic measurements for elastic modulus data.

1.4 LIBRARY OF CARBON MATERIALS USED IN THE NUCLEAR INDUSTRY

Characterizing other carbon-based materials commonly used in the nuclear industry will help to elucidate certain particular behaviors of nuclear graphite. In particular, understanding the irradiation response of carbon–carbon composites and glassy or pyrolytic carbon can help predict the performance of nuclear graphite.

The following articles and reports will support this section of the project:

1. "Microstructural characterization of the carbon-carbon composite JET tiles," journal article, status: in progress. Control samples used in the Joint European Torus fusion reactor are being used to understand the characterization of high-performance 2D composites that can be used for nuclear applications.

2. "Electron microscopy of the neutron irradiation effects of carbon–carbon FMI-222 composites," journal article, status: in progress. Unirradiated and neutron irradiated samples of FMI-222 composites were characterized to elucidate the irradiation effects on carbon fibers.

3. "Neutron irradiation effects in glassy carbon," journal article, status: in progress. Unirradiated and neutron-irradiated glassy carbon specimens underwent multimodal characterization. This research shows the effect of irradiation on this material and provides evidence that non-graphitizable material might not undergo the exact dimensional change known as turnaround in graphite.

2. FUTURE WORK AND LIBRARY REPOSITORY

Most of the data is currently stored in *the Mendeley scientific database* and *Data-in-Brief publications*. This data is planned to be eventually migrated to the *Gen IV Materials Handbook for Very High Temperature Reactor*.

The library data are also being processed to be used in future publications that will be incorporated into microstructural models that can be used to evaluate the performance of candidate graphite grades. The workflow and methodologies will be published in the form of journal publications.

3. SUMMARY

This project aims to create a comprehensive data library that describes graphite's key characteristics and microstructures. The project's strategy involves supporting the design, modeling, aging management, and decommissioning of graphite components. Efforts are also focused on understanding the uncertainty caused by microstructural variations in graphite. These variations can lead to differences in mechanical properties that must be considered in design and safety cases. Enhancing the characterization and understanding of irradiation effects in graphite production will facilitate the development of appropriate measures to address graphite variations.

The research focuses on the baseline characterization of unirradiated material, a guide to commonly used techniques for graphite characterization, exploration of defects caused by oxidation or irradiation, and an investigation of materials similar to graphite commonly used in the nuclear industry. Another key aspect of this research is to outline a protocol for assessing the consistency of grades and graphite components. The report also presents some preliminary results that contribute to understanding graphite's microstructure and aging mechanisms.

4. CITATIONS

[1] J.D. Arregui-Mena, N. Gallego, Report on initial development of a database of nuclear graphite characteristics based on microstructural characterization, Oak Ridge National Laboratory, Oak Ridge 2023.
[2] J.D. Arregui-Mena, P.D. Edmondson, D. Cullen, S. Levine, C. Contescu, Y. Katoh, N. Gallego, Microstructural characterization of the CGB graphite grade from the molten salt reactor experiment, Journal of Nuclear Materials 582 (2023) 154421.
[3] J.D. Arregui-Mena, D.V. Griffiths, R.N. Worth, C.E. Torrence, A. Selby, C. Contescu, N. Gallego, P.D. Edmondson, P.M. Mummery, L. Margetts, Using porous random fields to predict the elastic modulus of unoxidized and oxidized superfine graphite, Materials & Design 220 (2022) 110840.
[4] J.D. Arregui-Mena, D.V. Griffiths, R.N. Worth, C.E. Torrence, A. Selby, C. Contescu, N. Gallego, P.D. Edmondson, P.M. Mummery, L. Margetts, Microstructural characterization data of as received IG-110, 2114 and ETU-10 nuclear graphite grades and oxidation characterization data of IG-110, Data in Brief 44 (2022) 108535.
[5] J.D. Arregui-Mena, R.N. Worth, W. Bodel, B. März, W. Li, A.A. Campbell, E. Cakmak, N. Gallego, C. Contescu, P.D. Edmondson, Multiscale characterization and comparison of historical and modern nuclear graphite grades, Materials Characterization 190 (2022) 112047.
[6] J.D. Arregui-Mena, R.N. Worth, W. Bodel, B. März, W. Li, A. Selby, A.A. Campbell, C. Contescu, P.D. Edmondson, N. Gallego, SEM and TEM data of nuclear graphite and glassy carbon microstructures, Data in Brief 46 (2023) 108808.