Data Article

Microstructural characterization data of as received IG-110, 2114 and ETU-10 nuclear graphite grades and oxidation characterization data of IG-110

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A B S T R A C T

This manuscript provides optical microscopy, scanning electron microscopy, and transmission electron microscopy micrographs that show the microstructure of three superfine nuclear graphite grades IG-110, 2114 and ETU-10. This collection of microstructural data showcases the microstructure of these materials and helps to differentiate the most important features or phases of these graphite grades. In particular, the microstructural data illustrate the filler and binder

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morphology of these grades. Moreover, samples of as-received and oxidized IG-110 were characterized via optical microscopy and x-ray computed tomography. The microstructural data of oxidized IG-110 shows the porosity generated by oxidation experiments. These micrographs and data provide a unique insight into the microstructural features and oxidation effects in nuclear graphite and can be used to perform quantitative porosity analysis. This collection of microstructural data complements the modeling and characterization described in the associated manuscript, “Using porous random fields to predict the elastic modulus of unoxidized and oxidized superfine graphite (Arregui-Mena et al., 2022).”

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### Specifications Table

| Subject | Materials Characterization |
|---------|-----------------------------|
| Specific subject area | Material simulations and characterization of nuclear materials |
| Type of data | Tables – Descriptions of the selected graphite grades’ pores and other characteristics |
| | Figures – Micrographs in the form of transmission electron microscopy, scanning electron microscopy, and polarized optical microscopy |
| | Graphs – Plots showing the selected graphite grades’ pore size distributions |
| | - Transmission electron microscopy: JEOL JEM 2100F |
| | - Scanning electron microscopy: Tescan MIRA3 GMH SEM |
| | - X-ray computed tomography: Zeiss Xradia 620 Versa. |
| | - Optical microscopy: Nikon Microphot FXA and the Leica Application Suite software for unoxidized samples. Keyence digital microscope for oxidized samples. |
| Data format | The data were processed and included as images in the paper. |
| Parameters for data collection | - Transmission electron microscopy: JEOL JEM 2100F: acceleration voltage 200 kV |
| | - Scanning electron microscopy: Tescan MIRA3 GMH SEM: acceleration voltage 2 kV, beam current 8 μA |
| | - X-ray computed tomography: Zeiss Xradia 620 Versa: acceleration voltage 40 kV, power 5 W, current ~75 μA, source-sample distance: 12 mm, source-detector distance: 37.5 mm, 1601 projections with 13 s acquisition time. |
| Description of data collection | The data were collected using multiple characterization techniques, including optical microscopy, x-ray computed tomography, and scanning electron microscopy |
| Data source location | Institution: The US Department of Energy's Oak Ridge National Laboratory – Low Activation Materials Development and Analysis Laboratory |
| | City/State/Region: Oak Ridge, Tennessee |
| | Country: United States of America |
| Data accessibility | Included with the article and the raw data is provided in the following repository: |
| | Repository name: Mendeley Data |
| | Reserved DOI: 10.17632/c4sj6yctkd.2 |
| | https://data.mendeley.com/datasets/c4sj6yctkd |
| Related research article | J.D. Arregui-Mena, D.V. Griffiths, R.N. Worth et al., “Using porous random fields to predict the elastic modulus of unoxidized and oxidized superfine graphite, Materials & Design 220 (2022) 110840. |
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Value of the Data

- The micrographs and XCT data included in this manuscript illustrate the microstructure of three candidate graphite grades for molten salt reactors and high temperature reactors.
- This research will benefit the nuclear graphite community and the nuclear industry by characterizing the phases and by providing input data for modelling of porous random fields.
- The microstructural data will provide a deeper understanding of the differences among graphite grades and will help to inform microstructural base models.

1. Data Description

1.1. Microstructural Data of As-Received Graphite

This manuscript provides micrographs acquired via optical microscopy, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) that illustrate the microstructure of three candidate graphite grades (2114, IG-110, and ETU-10) for the future generation IV graphite-moderated reactors. The corresponding manufacturers of these grades are Mersen for 2114, ETU-10 for Ibaden and Toyo Tanso for IG-110, respectively. These manufacturers provided the samples used in this research. This article expands upon the research conducted in the article “Using porous random fields to predict the elastic modulus of unoxidized and oxidized superfine graphite” [1].

Even though 2114, IG-110, and ETU-10 might be considered similar graphite grades because they have a similar grain size and pore size distribution, they are made from different raw materials and have distinct pore networks. These sources of variability in the graphite materials make each grade unique. Microstructural and property variations are also expected across large components of graphite [2,3]. Overall descriptions of the phases of graphite and micrographs in Fig. 1 are provided below. Fig. 1 shows micrographs of 2114, IG-110, and ETU-10 graphite grades. Polarized optical microscopy revealed filler (F), binder (B), and porosity (P) regions. SEM micrographs show that the filler particles of different graphite grades have different shapes and textures. TEM micrographs illustrate the internal extructure of quinoline insoluble (QI) particles, cross sections of the binder and filler and thermal cracks.

**Filler particles** – This phase serves as an aggregate material. The most common origins of filler particles are petroleum, coal, or natural sources [4]. The bulk properties of nuclear graphite depend on the raw material source, orientation, and distribution of this phase [5]. Examples of filler particles in the selected graphite grades are shown in the optical and scanning electron microscopy (SEM) micrographs in Fig. 1. In the optical micrographs, the filler particle examples are located at the centers of the images. The SEM micrographs in Fig. 1 show differences, such as shape and texture, in these filler particles (delineated by dashed lines). The 2114 and ETU-10 filler particles were both manufactured from a nonpetroleum coke, resulting in a smooth monolithic structure, whereas the IG-110 filler particles were manufactured from a petroleum coke, resulting in multiple folded graphitic lamellae. Transmission electron microscopy (TEM) micrographs in Fig. 1 show other important aspects of the filler particles, thermal cracks, and crystal strains [6]. Additional information and data interpretation are found in refs. [7–11].

**Binder** – The binder phase initially consists of tars or pitches comprising a complex mixture of aromatic compounds [12]. Binder-rich areas around the filler particles are visible in the polarized optical micrographs in Fig. 1. The binder phase can be described as mosaic-like patterns formed by small domains with different colors caused by the random ordering of crystallites within this phase. The bottom section of Fig. 1 shows examples of cross sections of QI particles. It appears that the strength of the binder phase depends in part on the content of QI particles [13]. QI particles also play a role in the irradiation response of graphite as they undergo densification or fullerrenization [14,15].
Fig. 1. Polarized optical, SEM, and TEM micrographs of the selected graphite grades. Filler (F), binder (B), and porosity (P) features are indicated.
Fig. 2. Optical micrographs of unoxidized and oxidized samples of IG-110. WL – Weight loss.

The raw data in Fig. 1 can be found in an online repository: https://data.mendeley.com/datasets/c4sj6yctkd. A brief description of how these micrographs may be used and processed is described below:
Fig. 3. Reconstructed XCT data, segmentations of IG-110 unoxidized and uniformly oxidized samples, and open and closed porosity profiles across the data.
Polarized optical microscopy – These micrographs are often used to differentiate between filler particles and binder. These types of images are one of the tools used to measure the filler particle size in a grade as well as observe the binder domains in graphite.

SEM – This data provides a better understanding of the texture of filler particles and features contained in the binder, such as pores or thermal cracks.

TEM – Micrographs acquired via this technique illustrate fundamental differences between filler and binder.

1.2. Optical Microstructural Data of Oxidized Graphite

Optical micrographs of unoxidized and oxidized IG-110 samples are shown in Fig. 2. As can be seen from these micrographs, the oxidized samples have more and larger pores than those of the unoxidized specimen. The samples oxidized at 525 °C tend to be uniformly oxidized. By contrast, the samples oxidized at 750 °C had acute damage near the edge of the sample. Regime 1 tends to evenly oxidize graphite specimens, while Regime 3 will preferentially remove material from the edge of the surface. Regime 1 (low temperatures) results in a more even and extended damage and therefore has a more significant impact on the mechanical performance of a graphite component than the acute oxidation in Regime 3 (high temperatures) which is limited to the surface of a graphite specimen. Regime 2 oxidation produces more complex microstructural changes across the sample [16]. Data contained in Fig. 2 is stored in the online repository: https://data.mendeley.com/datasets/c4sj6yctkd. Segmentation and analysis of this data can help understand the oxidation profiles and microstructural changes induced by different oxidation regimes. In particular, the optical micrographs can be used to compare how each oxidation regime affects the microstructure and pore size distribution of oxidized IG-110.

1.3. X-ray Computed Tomography of Oxidized Graphite

To further characterize the damage produced by chronic oxidation, some IG-110 samples were characterized using x-ray computed tomography (XCT). Fig. 3 shows XRT images of two uniformly oxidized samples and one unoxidized sample along with plots showing open and closed porosity content across the segmented data. The plots and different illustrations included in Fig. 3 illustrate the effects of uniform oxidation across the samples. The plots presented in Fig. 3 are the first visual representation of the effects of oxidation in graphite using a 3D characterization technique. The 2D slices and 3D reconstructions both show the oxidation across the microstructure in IG-110.

2. Materials and Methods

2.1. Materials

Graphite was the moderator material of the first nuclear reactors, and it is used or being proposed for use in the current fleet of British advanced gas-cooled reactors (AGR) and in generation IV concepts such as the very high temperature reactors (VHTRs) and molten salt reactors (MSRs) in the United States. The list of materials and general information on each graphite grade is given in Table 1.

2.2. Metallographic Preparation of Optical Microscopy Samples

To image the specimens, coupons of each graphite grade and oxidized samples were embedded in epoxy resin and mechanically polished. Mechanical polishing was performed using
Table 1
List of surveyed materials and description of selected graphite grades.

| Graphite grade | Coke source          | Forming process | Type     | Grain size (μm) | Manufacturer | Application             | Ref.       |
|----------------|----------------------|-----------------|----------|-----------------|--------------|--------------------------|-----------|
| 2114           | Nonpetroleum coke   | Isostatically molded | Superfine | 13              | Mersen       | Gen IV reactors          | [17,18]   |
| ETU-10         | Coal-tar pitch      | Isostatically pressed | Superfine | 15              | Ibiden       | Gen IV reactors          | [19]      |
| IG-110         | Petroleum coke      | Isostatically pressed | Superfine | 20              | Toyo Tanso   | Gen IV reactors          | [17,18]   |

Table 2
Oxidation parameters and techniques used to characterize IG-110 specimens.

| Techniques                  | Oxidation temperature (°C) | Weight loss (%) |
|-----------------------------|----------------------------|------------------|
| Optical microscopy/XCT      | Unoxidized                 | —                |
| Optical microscopy/XCT      | 525                        | 4.5              |
| Optical microscopy/XCT      | 600                        | 5                |
| Optical microscopy          | 750                        | 5                |

Fig. 4. Sample polishing preparation process.

a Sruers TegraFroce-5/TegraPol-31 polishing system. The steps used to polish the samples are listed in Fig. 4.

2.3. Oxidation Experiments

All oxidation tests were performed on fresh specimens extracted from billets of IG-110. In preparation for tests, the specimens were cleaned and dried, and then their mass and dimensions were measured. Oxidation tests were performed at the temperatures listed in Table 2. The specimens were recovered after oxidation, and their mass and dimensions were measured again. The oxidation apparatus is in a laboratory with central air conditioning that maintains an environment with a stable temperature and constant humidity.
2.4. Optical Microscopy Image Acquisition and Processing

The images of polished unoxidized samples were captured with an optical microscope (Nikon Microphot FXA) and the Leica Application Suite software. The optical texture of the filler, porosity, and binder phases were used to identify each graphite phase. A Keyence digital microscope was used to capture the optical micrographs of the polished oxidized specimens.

2.5. X-Ray Computed Tomography Acquisition and Processing

To illustrate the effects of temperature and different oxidation regimens, two IG-110 samples oxidized in an air environment were characterized using XCT and optical microscopy. A special segmentation methodology was implemented to differentiate between the open and closed porosity. A summary of the segmentation methodology is given in Fig. 5. The first step consists of segmenting the porosity from the reconstruction and creating the label for the overall porosity. The Otsu criterion [20] was used to differentiate between the porosity and the solid phase. Next, a layer of voxels, an outside label, is drawn surrounding the volume of interest to differentiate between the outside and the border of the reconstruction. Then, the individual pores or network of pores connected with the outer label are assigned as open porosity. The remaining pores or network of pores are assigned as closed porosity. This methodology attempts to replicate the definitions of open and closed porosity commonly used in nuclear graphite research.

Ethics Statement

No ethics conflicts are associated with this publication; this research involves only the study of materials.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships which have or could be perceived to have influenced the work reported in this article.
Data Availability

Microstructural characterization data of as received IG-110, 2114 and ETU-10 nuclear graphite grades and oxidation characterization data of IG-110 (Original data) (Mendeley Data).

CRediT Author Statement

José David Arregui-Mena: Data curation, Methodology, Visualization, Writing – original draft; D.V. Griffiths: Writing – review & editing, Software; Robert N. Worth: Investigation; Christa E. Torrence: Writing – review & editing; Aaron Selby: Writing – review & editing; Cristian Contescu: Methodology, Writing – review & editing, Conceptualization; Nidia Gallego: Writing – review & editing; Philip D. Edmondson: Writing – review & editing; Paul M. Mummery: Writing – review & editing; Lee Margetts: Writing – review & editing, Software.

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