Nitrogen adsorption data, FIB-SEM tomography and TEM micrographs of neutron-irradiated superfine grain graphite

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This manuscript provides raw nitrogen gas adsorption data, images and videos obtained from a technique that combines Focused Ion Beam (FIB) and Scanning Electron Microscopy (SEM) known as FIB-SEM tomography and Transmission Electron Microscopy (TEM) micrographs. This collection of data is useful for characterization of the effects of high fluence neutron irradiation in nuclear graphite as described in the associated manuscript, “Mesopores development in superfine grain graphite neutron-irradiated at high fluence” (Contescu et al., 2019). Nitrogen adsorption isotherms at 77 K are provided for graphite samples before and after neutron irradiation at 300, 450, and 750 °C at fluences before and after turnaround. FIB-SEM tomography reveals porosity of unirradiated and irradiated samples. Using this technique, four data sets were obtained, of which the first three are presented in video format, whereas the fourth one is a series of images provided in raw format unique to this manuscript. All microscopy data document the microstructure, surface area and...
porosity of superfine grain graphite G347A (Tokai Carbon, Japan) before irradiation and irradiated after turnaround at 400 °C. TEM micrographs provide unique information on irradiation damage at high neutron fluence (> 27.8 displacements per atom, dpa) in the microstructure and crystal lattice of graphite. Additional TEM micrographs are provided here, which do not duplicate the research paper published elsewhere (Contescu et al., 2019). These data sets are unique, as samples at high irradiation doses have never been measured or imaged before with the aforementioned techniques.

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**Specifications table**

| Subject area          | Material science |
|-----------------------|------------------|
| More specific subject area | Nuclear engineering, Microscopy, Nuclear Materials |
| Type of data          | Gas adsorption isotherms data, TEM images and FIB-SEM tomography slices in video format |
| How data was acquired | Data was obtained using an Autosorb 1C instrument (Quantachrome), a FEI Versa 3D Dual beam and a JEOL JEM 2100F TEM at an acceleration voltage of 200 kV. |
| Data format           | Raw data of nitrogen adsorption isotherms (Excel format), TEM micrographs (dmr files) and SEM images (tiff format) |
| Experimental factors  | In the case of TEM bulk samples where polished to obtain flat surfaces, after these lamellae where obtained with FIB preparation techniques |
| Experimental features | Equilibrium adsorption isotherms were obtained using N2 at 77 K, the weight of specimens were 0.53 g. FIB-SEM data where obtain to create 3D models of the porosity of graphite. TEM micrographs were taken from foils to analyze the irradiation effects and changes produced by neutron irradiation. |
| Data source location  | The data was collected at Oak Ridge National Laboratory’s Low Activation Materials Development and Analysis (LAMDA) located at Oak Ridge, Tennessee, US |
| Data accessibility    | Data is with this article. |
| Related research article | Mesopores development in superfine grain graphite neutron-irradiated at high fluence, [1] |

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**Value of the data**

- Detailed gas adsorption isotherms with a total of 117 points can be used to re-examine the BET surface area and pore size distribution modified by neutron irradiation
- TEM data shows several examples of neutron irradiation damage on the crystal structure of graphite at high doses. TEM micrographs can be used to measure the size and study the morphology of cracks found in neutron irradiated samples
- Fast Fourier Transform (FFT) analysis can be performed on TEM micrographs to quantify the degree of crystallinity of the neutron irradiated samples FIB-SEM tomography data can be used to quantify the pore volume fraction of the given SEM data sets
- TEM images allow other researchers to study the crystal structure and porosity structure of irradiated graphite
1. Data

The superfine grain graphite grade G347A used in this research was manufactured by Tokai Carbon Co., Ltd. All the irradiated samples used in this research were irradiated at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL).

1.1. Gas adsorption

A series of equilibrium adsorption/desorption isotherms were measured with the Autosorb 1C instrument (Quantachrome) using N2 at 77 K. Most of the specimens were outgassed for 2 to 3 h at 300 °C prior to measurements, only the samples irradiated at 300 °C were outgassed at a lower temperature (265 °C) to prevent annealing of irradiation damage during outgassing. In total 117 data points were acquired during adsorption and desorption processes. Table 1 shows the irradiation conditions for the specimens characterized with this technique.

1.2. FIB-SEM tomography data

A microscopy technique that combines Focused Ion Beam (FIB) and Scanning Electron Microscopy (SEM) known as FIB-SEM tomography was used to scan a total of four regions. FIB-SEM tomography is capable of obtaining serial high-resolution SEM images of a section of a specimen as is described in reference [2].

The description of the samples where the scans were performed are as follows: an unirradiated sample and two specimens exposed to high neutron doses. These data sets are provided in different formats: the data used for the associated publication [1] is given in a video format that shows SEM images recorded continuously at higher and higher depths inside the specimen during the FIB operation. In contrast, only a few frames of these continuous videos were provided in the associated publication [1]. The fourth set of FIB-SEM data, unique to this publication, is given in an image raw format (Fig. 1). The information of images and conditions of the fourth data set are summarized in Table 2. A FEI Versa 3D Dual beam was used to acquire the images at an acceleration voltage of 5 kV.

| Nominal Irradiation temperature °C | Fluence $E > 0.1$ MeV $10^{25}$ n/m$^2$ | dpa | Comments |
|-----------------------------------|------------------------------------------|-----|----------|
| None                              | None                                     | None| Un-irradiated reference |
| 300                               | 22.8                                     | 16.6| Turnaround |
| 450                               | 40.8                                     | 29.8| Maximum swelling, still $\Delta V/V < 0$ |
| 600                               | 12.6                                     | 9.2 | Before turnaround |
| 40.8                              | 21.0                                     | 15.3| Turnaround |
| 750                               | 27.8                                     | 20.3| Past turnaround, still $\Delta V/V < 0$ |
| 13.8                              | 36.1                                     | 26.4| Initial swelling, $\Delta V/V > 0$ |
| 23.8                              | 40.8                                     | 29.8| Maximum swelling |
| 29.2                              | 17.8                                     | 13.0| Dose comparison (*) |
| 23.8                              | 7.9                                      | 5.8 | Before turnaround (*) |
| 13.8                              | 13.8                                     | 10.1| Turnaround |
| 29.2                              | 23.8                                     | 17.4| Dose comparison $\Delta V/V \approx 0$ |
| 1.4                               | 29.2                                     | 21.3| Maximum swelling |

Note: Gas adsorption was not performed on specimens marked with asterisks (*)

Table 1

Irradiation conditions and properties measured by gas adsorption.
1.3. TEM data

Transmission Electron Microscopy (TEM) micrographs were only produced for the irradiated samples. The irradiation conditions of these samples are given in Table 3. The TEM foils used in this research were obtained using FIB sample preparation techniques. The TEM micrographs provided

Fig. 1. Raw data of FIB-SEM tomography images obtained from a specimen irradiated at 26.4 dpa and 400 °C (selected slices).

| Sample | Nominal irradiation temperature (°C) | Number of slices/images | Pixel resolution raw data (nm) | Slice thickness (nm) |
|--------|------------------------------------|-------------------------|-------------------------------|---------------------|
| 26.4 dpa | 450                    | 365                      | 24                           | 15                  |

Note: dpa = displacements per atom

Table 2
Irradiation conditions and information of the SEM images for the raw data set.

Table 3
Number of TEM images and conditions for each irradiated sample.

| Sample | Nominal irradiation temperature (°C) | Number of images |
|--------|------------------------------------|------------------|
| 15.3 dpa | 450                        | 12               |
| 26.4 dpa | 450                        | 9                |
| 29.8 dpa | 450                        | 14               |
| Total   |                              | 35               |

Note: dpa = displacements per atom

1.3. TEM data

Transmission Electron Microscopy (TEM) micrographs were only produced for the irradiated samples. The irradiation conditions of these samples are given in Table 3. The TEM foils used in this research were obtained using FIB sample preparation techniques. The TEM micrographs provided
were taken with a JEOL JEM 2100F TEM at an acceleration voltage of 200 kV. TEM micrographs presented here are unique to this manuscript and are given in Figs. 2–5. The original files for the associated paper are also given in a dm3 format.

Figs. 2 and 3 show some of the features found in the same TEM foil prepared from one of the irradiated specimens (15.3 dpa). The first compilation of images (Fig. 2) illustrates a series of images taken from the location indicated in the individual images (Fig. 2a-b). The first set of images compiled in Fig. 2 shows high contrast areas in the crystal structure of graphite. In Fig. 3 it can be observed the “feather” like contrast around the cracks.

Fig. 2. Features found in the sample irradiated at 15.3 dpa.

Fig. 3. Feather like contrast around cracks of sample irradiated at 15.3 dpa.

Fig. 4. Features found in the sample irradiated at 29.8 dpa.

Fig. 5. Features found in the sample irradiated at 29.8 dpa.
Turning now to the sample irradiated at 29.8 dpa, TEM images reveal the complex structure of pores (Fig. 4) and the possible effect of neutron irradiation in quinoline insoluble (QI) particles. In Fig. 5a can be observed densification of quinoline insoluble (QI) particles after neutron irradiation, as proposed also by Karthik et al. [3]. Moreover, Fig. 5b shows cracks around the QI particles that may have been produced by the densification of these particles.

2. Experimental design, materials, and methods

2.1. Experimental design

Details on the origin of graphite specimens, their irradiation conditions, and post-irradiation examination are available somewhere else [3]. A subset of specimens irradiated at three temperatures (300, 450 and 750 °C) and fluxes up to 30 dpa, covering stages before and the turnaround point, was selected for further characterization.

Nitrogen adsorption isotherms at 77 K (with 117 points on adsorption and desorption branches) were measured for all irradiated samples in this subset and for an unirradiated reference sample. These data are provided in Excel format, and can be used to extract or re-analyze the main texture properties of irradiated graphites (BET surface area, DFT pore size distribution, BJH pore size distribution etc) and to compare with dimensional changes reported elsewhere [1,4].

Three electron microscopy techniques (SEM, TEM, FIB-SEM) were used for examination of porosity and microstructure of specimens irradiated at 450 °C. The experimental details on samples preparation for SEM, TEM and FIB-SEM are presented next.

2.2. General preparations - Polishing of the samples

A flat surface is preferable to optimize the FIB-SEM tomography acquisition processes and TEM lift-out preparation, therefore each sample was mechanically polished. In this case the specimens were mounted in epoxy material to improve the handling and reduce possible contamination of the instrument.

2.3. FIB-SEM tomography sample preparation and method

FIB-SEM tomography is a technique that can be used to analyze the complex microstructure of graphite at high resolutions (~10 nm) [5]. This technique consists in using a dual beam Scanning Electron Microscope (SEM) and Focused Ion Beam (FIB) instrument to mill and image an area of interest (Fig. 6).

In order to start the sample preparation, eucentric height (the position where the electron and ion beam coincide) is achieved by adjusting the position of the stage and beams. After eucentric height is obtained the sample is tilted to a 52° angle for FEI systems and start the pre-milling/imaging operations. The general set-up for the FEI Versa Dual Beam is shown in Fig. 7.

Several important elements conform the pre-milling/imaging operations: a protective platinum layer, a fiduciary mark, milling direction and trenches around to the area of interest. All these elements can be visualized in Fig. 8.

- **Application of the platinum layer** – this step involves covering the area of interest with a layer of platinum that serves as a protective material from ion beam damage.
- **Fiduciary mark milling** – A layer of platinum deposited close to the area of interested followed by the milling of a fiduciary mark. This mark is used to track drift and working distance by the software of the Dual FIB-SEM instrument.
- **Milling direction** – The arrow in Fig. 7 indicates the direction in which the sample will be milled for imaging the exposed surface
Fig. 6. Sequential imaging and milling processes of FIB-SEM tomography.

Fig. 7. FIB-SEM dual beam components and sample position for FIB-SEM tomography.

Fig. 8. Preparation features for FIB-SEM tomography.
• **Milling of trenches** – Normally three trenches are milled around of the area of interest, these trenches provide access to the surface image for the SEM beam, reduce shadowing effects during the imaging and reduce possible redeposition of material.

### 2.4. TEM sample preparation and characterization

TEM samples were prepared with standard FIB sample preparation from mechanically polished samples, low voltages and currents were used for the lift-out procedures and thinning of the lamellas to minimize ion damage on the samples. The specimens were welded to a standard TEM grid. The JEOL 2100F located at ORNL’S LAMDA laboratory was used at an acceleration voltage of 200 kV in TEM mode at room temperature.

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### Transparency document. Supporting information

Transparency data associated with this article can be found in the online version at https://doi.org/10.1016/j.dib.2018.11.078.

### Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at https://doi.org/10.1016/j.dib.2018.11.078.

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