X-ray tomography for micro-structural characterization of fusion technology relevant composite materials

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Abstract
The evaluation of total porosity and its parameters as open porosity, pore area or size distribution are important factors in evaluation and selection of carbon-based materials - carbon-fiber composites (CFC) and graphite - for the projects in fusion energy production. The techniques used to measure the porosity of materials have very much evolved in diversity and performance, from gas adsorption/permeation or liquid extrusion to direct pore imaging. Until recently only imaging technologies providing sufficient resolution to reveal directly pores in size ranges that correlate with the mechanical, thermal or other physical properties relevant for the uses of CFCs as construction materials have been used. This approach has the merit of being flexible (a given image can be processed repetitively with e.g. newer algorithms better adapted for the various porosity characteristics). On the disadvantage side, one image characterizes only a thin slice of investigated material and thus may not be representative for the whole sample that can present important inhomogeneities or anisotropy. In the particular case of CFCs, which have an important structural anisotropy, it is necessary to image and process a big number of slices (sections) with many spatial orientations. With the advent and fast evolution of micron-resolution X-ray tomography in the last few years, the possibilities of high-resolution digital image processing were complemented with the advantage of the image data acquisition in the whole volume of the sample, during a single exposition to X-rays. Here we have sustained an intense campaign of X-ray microtomography measurements of most relevant composite materials for fusion technology measured in the same conditions. To our knowledge, this is the first time when such consistent comparison of different composite materials is proposed. In our method the porosity factor is determined as the ratio between the area of the difference between the measured histogram and the symmetrized histogram with the help of a generalized Gaussian fit. We show that our procedure for the quantitative evaluation of the sample porosity factor yields realistic results for the three types of CFC analyzed.

Keywords: CFC, porosity factor, X-ray microtomography

1 Introduction
The carbon-carbon fiber reinforced (CFC) materials offer attractive properties for plasma facing components (PFC) in future fusion power plants and for the thermal protection system of space vehicles. The carbon reinforced carbon fiber (CFC) monoblocks of ITER diverter vertical target must sustain high heat fluxes of 10 MW/m² during 400 s (normal operation) and up to 20MW/m² during 10 s (off-normal event). CFCs have a unique combination of high conductivity, low Z and resistance to damage
induced by the high heat loads. Given the demanding environmental requirements of ITER diverter, specially developed CFCs are needed as plasma facing components (PFC) materials. The problem of fuel (tritium) retention in a carbon material is a major concern due to its radioactivity and inventory constraints within the ITER installation. The spatial distribution of porosity and density of these materials has to be determined for assessing their performance at high temperatures and/or intense irradiation fields. The major role of porosity in both fabrication and operating of PFCs is induced by the (i) the quality of actively cooled components which depends on the metal impregnation inside the macro-pores of the composite and (ii) the co-deposition mechanism inside CFC porosity which is responsible for the most of the fuel retention. It is expected that an accurate 3D porosity description of the CFC materials provides an essential input for the quantization of the fuel retention in the material bulk.

3D X-ray computed micro-tomography (µXCT) is a convenient technique used to monitor the morphology on extended volumes. The method can reveal the occurrence, distribution and shape of the regions with a different density at macro local scale. µXCT is the only tool available for the simultaneous determination of porosity factor and for the 3D visualization of the pore network on a rather macroscopic sample (up to 5x5x5 mm$^3$).

2 Materials and methods
The non-invasive inspection was pursued on samples with or without refractory/marker metal coating, non-irradiated or post-mortem samples. The main challenge is posed by the required micron range of the spatial resolution for rather macroscopic samples.

Most experiments were carried out at our newly upgraded X-ray tomography facility [1] (Fig.1). The system is equipped with a last generation Nano-focus X-Ray source for non-destructive inspection. The source is operational in both micro- and nanofocus mode, at a tube voltage up to 225 kV and a maximum power of 10÷20 W. X-Ray images can be acquired by using three different high resolution detector types: Image Intensifier (XII) for rapid non-destructive examinations, flat panel (FP) detectors and a linear detector for the high density sample analysis. Positioning and turning around of the sample are insured by a set of seven high precision motorized micrometric manipulators.

![View of the NILPRP Nano-CT facility.](image-url)

Figure 1: View of the NILPRP Nano-CT facility.
Micro-focus X-Ray source
Max. high voltage: 225 kVp
Feature recognition: <1 µm
Min object-focus distance: 0.3 mm
X-Ray cone: 170°
X-ray target power: <10÷30 W

X-Ray detection:
- Detector elements/effective area
  - 768x576 / 169x169 mm² 7” XII
  - 1024x1024/ 50x50 mm² CMOS FP
  - 1944x1536/ 145x115 mm²
  - a-Si FP panel sensor 1024x1024 flat panel sensor

Digital Output
10 to 16 bits

Sample micrometric manipulator
- X stage
  - travel up to 800 mm, loading capacity up to 30 kg
- Z stage
  - travel 300 mm, loading capacity up to 6 kg
- θ stage
  - accuracy 0.03°, loading capacity up to 10 kg

Magnification factor
< 2000

Spatial resolution
≥ 500 nm (JIMA mask)

Scanning time
< 15 min. (1000 views)

3D reconstruction time
< 15 min (2048x2048x1024 voxels)

Scan method
Cone Beam CT; short scan CT (180° + fan angle) and Oblique cone-beam CT

Table 1 - X-Ray micro-tomographic system parameters

Automation, control and data acquisition were obtained by means of in-house software package. The tomographic reconstruction for the cone-beam scanning is based on an optimized implementation of the modified cone beam filtered back-projection algorithm. Using a parallelization technique on multiprocessors workstations, experimental data consisting of large radiographic images (1944x1536 pixels) are processed for building the 3D reconstructions of typically 2048x2048x1024 voxels. The most important characteristics of the X-Ray tomographic system are presented in Table 1. We have sustained an intense campaign of CT measurements of most relevant composite materials for fusion technology. For each type of material we cut three samples which have been measured in the same conditions. To our knowledge, this is the first time when such consistent comparison of different composite materials is proposed.

Optimization of the X-ray tomography setup in order to obtain appropriate spatial resolution for the analysis of the composites structure has been carried out. Materials scientist is interested in achieving a feature recognition value in the range of couple of microns. It is already a standard in our laboratory to use the JIMA mask as space resolution benchmark. The feature recognition of the X-ray source – detector system was continuously checked. Figure 2 shows a typical radiography of the JIMA mask in the micro-focus mode. One can see that the system can regularly achieve a detail recognition value under 3 µm. A set of 1440 radiographies at equidistant angles have been used for the high resolution fully 3D tomography. The voxel size was kept at 3.5 µm for the inspection of samples of 4x4 mm² cross-sections.
In the current work we apply cone beam micro-tomography to the comparative characterization of CFC materials relevant for fusion technology: NB31 and NB41 (SEP/Snecma) and DMS780 (Dunlop). For reference, homogenous fine-grain graphite (FGG) EK98 samples were also scanned. Table 2 gathers the materials and the relevant fusion facilities.

<table>
<thead>
<tr>
<th>Material</th>
<th>Manufacturer</th>
<th>Fusion facility</th>
<th>Density [g/cm³]</th>
<th>Open porosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>CFC NB41</td>
<td>SEP/Snecma</td>
<td>ITER</td>
<td>&gt;1.94</td>
<td>&lt;7%</td>
</tr>
<tr>
<td>CFC NB31</td>
<td>SEP/Snecma</td>
<td>Former ITER</td>
<td>1.87-1.93</td>
<td>7%-9%</td>
</tr>
<tr>
<td>CFC DMS780</td>
<td>Dunlop</td>
<td>JET</td>
<td>1.75 – 1.87</td>
<td>12% - 15%</td>
</tr>
<tr>
<td>FGG EK98</td>
<td>Ringsdorff</td>
<td>TEXTOR</td>
<td>1.85</td>
<td>≈ 10%</td>
</tr>
<tr>
<td>CFC N11</td>
<td>SEP/Snecma</td>
<td>Tore Supra</td>
<td>1.71 – 1.78</td>
<td>≈ 12%</td>
</tr>
</tbody>
</table>

Table 2: Carbon based composite materials of fusion technology relevance [2]

2.1 Qualitative comparison of CFC morphologies

Figures 3 a, b illustrate the morphology of JET type DMS780 CFC, a 2-D CFC formed by planes of PAN (polyacrylonitrile) fiber bundles perpendicularly oriented with interlayers (felt) of PAN fibers somehow randomly oriented. The size of the sample cross-section is ≈ 4 mm. The minimum detectable feature is a single fiber of 6-7 µm. The PAN fibers from the felt have a cladding of graphite of around 20 µm radius.

Figure 3: a) 3D tomography reconstruction with the main components; b) the elementary module composed of one PAN and two felt layers.
Figures 4 show the architecture of the NB41, a state-of-the-art three-dimensional type of CFC.

![Figure 4: 3D tomography reconstruction with the main components](image)

The manufacturer considers that the two materials NB31 and NB41 are very similar 3D CFC types. However, one can see in Figure 5 some morphology differences related especially to the somehow more ordered nature of the newly developed NB41. Thus NB41 has in general thinner pitch fiber layers with a clearer grid structure. Also the proportion of pitch fibers oriented in Z direction (horizontal in the picture) is larger.

![Figure 5: Morphology differences between two types of 3D structured CFCs: NB31 and NB41. Note the somehow more ordered nature of the newly developed NB41.](image)

### 2.2 Determination of porosity factors

The evaluation of total porosity and its parameters as open porosity, pore area or size distribution are important factors in evaluation and selection of carbon-based materials - carbon-fiber composites (CFC) and graphite - for the projects in fusion energy production. Since the first fusion experiments, nearly fifty years ago, the techniques used to measure the porosity of materials have very much evolved in diversity and performance, from gas adsorption/permeation or liquid extrusion to direct pore imaging. The porosity measurements for carbon materials used in projects currently running or nearing commissioning were regularly done by processing 2D gray-level images of plane sections cut from CFC samples on various directions relative to their fiber structure [3-5]. The images were obtained by optical or electron transmission microscopy or by micro-radiography. Until recently these methods were the only imaging technologies providing sufficient resolution to reveal directly pores in size ranges that correlate with the mechanical, thermal or other physical properties relevant for the uses of CFCs as construction materials. Simultaneously, the X-ray tomography of enhanced resolution of the order of 10 started to be employed in the investigation of relatively large porosity [6-7].
This approach has the merit of being flexible (a given image can be processed repetitively with e.g. newer algorithms better adapted for the various porosity characteristics). On the disadvantage side, one image characterizes only a thin slice of investigated material and thus may not be representative for the whole sample that can present important inhomogeneities or anisotropy.

In the particular case of CFCs, which have an important structural anisotropy, it is necessary to image and process a big number of slices (sections) with many spatial orientations, to fully characterize the sample. This is obviously expensive in terms of working effort and time required to make a complete porosity characterization for a given sample.

With the advent and fast evolution of micron-resolution X-ray tomography in the last few years, the possibilities of high-resolution digital image processing were complemented with the advantage of the image data acquisition in the whole volume of the sample, during a single exposition to X-rays [1, 8]. Of course, the presence of specific artifacts in X-ray tomographs may require some validation by classical methods as mercury [9] or gas porosimetry [5, 10], that reveal mainly the open porosity.

The tomography analysis is able to provide useful quantitative information about the porosity network of the composite samples and can be used to identify the relevant mechanism for fuel retention into the material bulk like: fuel localization in the bulk or in the trapping sites (porosity) along fibers, or co-deposition into CFC open pores.

Figure 6 shows the attenuation coefficients histograms of the whole set of measurements. The measurements were carried out in the same conditions (X-ray energy and intensity, filters, magnification etc). The focus spot of the X-ray tube was benchmarked with the JIMA mask before each measurement. The densities of the materials are very well reproduced by the tomographic measurements. The histograms of NB31 and NB41 are very similar, as they are considered by the manufacturer to be almost identical materials. The 2D Dunlop manufactured DMS780 material has a lower density and clearly higher porosity.

![Figure 6: Histograms of the attenuation coefficients for fusion technology relevant composite materials and a homogenous non-porous material.](image)

A procedure for a quantitative evaluation of the sample porosity factor has been recently introduced and tested in [1]. The post-processing of the reconstructed voxel volume comprises several steps: (1) finding the optimal choice for the threshold level for voxel values (attenuation); (2) following validation, the reconstructed volume is segmented and the porous structure is extracted as an independent object which can be represented also as a 3D structure; (3) determination of the absolute value of the porosity factor. Voids volume, size and projected area distribution can also be determined. With this method we obtained porosity factors for all fusion technology relevant CFC materials, in good agreement with the manufacturer specifications. The porosity factor uncertainty was evaluated.
based on the scattering of the estimated values for different samples of the same material and/or for different region of interest within the reconstructed volume. The uncertainty so determined is around half of a percent.

It must be stressed that, due to partial volume effect (the voxels located on pore edges have intermediary values between the attenuation of the sample material and that of voids), the information about porosity is easy to extract only for pore sizes sensibly larger than the resolution of tomography. This problem is treated in literature for different tomographic techniques, pore topologies and size distributions [11-12], and one can expect further improvement in this field.

The porosity factor was also evaluated by means of a second method. To demonstrate that the asymmetry of the attenuation coefficients histogram is related to the porosity of the material we present a simple experiment – tomography of two materials, with and without pores. Figure 7 shows a CT cross-section comparing a non-porous material (Teflon) with fine grain graphite EK 986, scanned side-by-side.

![Figure 7: μXCT of a non-porous material (Teflon) and of FGG EK986 (bottom) which has rather small pores.](image)

The histograms of the non-porous sample (Teflon) and of FGG EK986 are presented in Figure 8 a, b. As expected, the Teflon associated histogram is symmetric with a perfect fitting Gaussian. The histogram for FGG is clearly asymmetric as shown in Fig 8b.

![Figure 8: a) Symmetric histogram (blue) of a non-porous material (Teflon) and the Gaussian fit (red), b) histograms for Teflon (red) and EK 986, asymmetric (green)](image)

The method for porosity evaluation would make use of this difference in the density profiles as follows:

- in a 3D visualization program (for example Volume Graphics, http://www.volumegraphics.com) a rectangular 3D selection is made;
- the histogram of the selected volume is generated;
- the histogram is further processed by fitting the right half (higher grey values) with a generalized Gaussian function;
- finally, the porosity factor is determined as the ratio between the area of the difference between the measured histogram and the Gaussian fitted one and the area of the Gaussian. The last step is illustrated in Figure 9 a,b. The porosity factor associated with this figure is 6.4% for NB41 CFC and 12.5% for the more porous 2D CFC DMS780.

![Image](image-url)

Figure 9: The porosity factor is determined as the ratio between the area of the difference between the measured histogram (blue) and the Gaussian fitted one (red) and the area of the Gaussian. a) NB41, b) DMS780.

The porosity factors are in good agreement with the manufacturer specifications. One can note that the method accounts very well for the excessive porosity of the 2D-structured DMS780 CFC material. It is also clear that in the case of EK 986 one underestimate the porosity due to the limited special resolution of the μXCT method.

<table>
<thead>
<tr>
<th>Material</th>
<th>NB31</th>
<th>DMS780</th>
<th>N11</th>
<th>NB41</th>
<th>EK 986</th>
</tr>
</thead>
<tbody>
<tr>
<td>Porosity factor (%)</td>
<td>6.8</td>
<td>12.5</td>
<td>10.5÷12</td>
<td>6.4</td>
<td>4.25</td>
</tr>
</tbody>
</table>

Table 3: Porosity factors evaluated from tomography reconstructions

Using the porosity factor procedure one can determine the overall porosity factor of a composite material (for example the DMS780 CFC) as a combination of the relatively low porosity of the PAN fiber layers (6÷8 %) and the more felt regions (≥20%).

3 Conclusions

Micro-tomography analysis was used for the 3D modeling of the fusion technology relevant CFC materials. For each type of material we cut three samples which have been measured in the same conditions. To our knowledge this is first time that such consistent comparison of different composite materials has been done. High resolution morphology (voxel size=3.5 µm) of rather macroscopic CFC samples (4x4x4 mm³) was obtained. We show that our procedure for the quantitative evaluation of the sample porosity factor yields realistic results for the three types of CFC analyzed. The results obtained by 3D micro-tomography analysis of statistically relevant volumes of CFC can be considered as a good basis for the characterization of the initial porosity of the new CFC ITER reference material NB41.
References