MULTI-SCALE QUANTITATIVE ANALYSIS OF CARBON STRUCTURE AND TEXTURE: III. LATTICE FRINGE IMAGING ANALYSIS

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Introduction

The various transmission electron microscopy modes provide textural and structural information on carbon materials at a wide range of scales (e.g., [1, 2]). In particuler, high resolution, 002 lattice fringe (LF) images are the only means to directly visualize the graphene planes arrangement, i.e., the sample nanotexture.

We present herein the methodology for LF image analysis we developed, then implemented using the Python scripting language, enabling us to extract several quantitative nanotextural parameters. Our method was then applied to the study of the texture of the dense pyrocarbon layers in several TRISO fuel particle samples.

Background

The recent years have seen a renewed interest in the high temperature reactor (HTR) technology for nuclear power plants [3]. In such a reactor, the fuel is composed of small ($\sim$1 mm) spherical particles, with a fissile or fertile uranium core and a multilayer protective coating (Fig. 1).

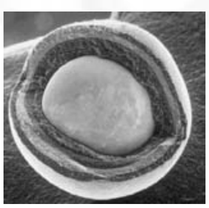

Figure 1: Exemple of TRISO HTR fuel particle. The layers are, from the kernel out: porous pyrocarbon (buffer), inner dense pyrocarbon (IPyC), SiC, outer dense pyrocarbon (OPyC).

This coating is deposited by fluidized bed chemical vapor deposition (CVD) and is composed of the following layers: *(i)* a porous pycrocarbon layer (buffer); *(ii)* an inner dense pyrocarbon layer ("IPyC"); *(iii)* a silicon carbide (SiC) layer; *(iv)* an outer dense pyrocarbon layer ("OPyC") [4].

Each layer has a specific function. The buffer absorbs the gaseous fission products and allows for mechanical expansion of the core. Fission products confinement is mainly performed by the SiC layer. The IPyC layer protects the core from the corrosive chemicals used during SiC deposition, and the OPyC layer protects the SiC layer.

The dense pyrocarbon (PyC) layers also act as a mechanical reinforcement for the SiC layer: as neutron irradiation induces an increase in anisotropy and a (anisotropic) densification of the PyC layers, they apply a compressive stress on the SiC, thus counteracting the outward pressure from fission products.

However, if irradiation-induced PyC densification is too important, structural weaknesses may appear and propagate to the SiC, as the PyC layers are mechanically coupled to the SiC layer, and cause confinement failures.

Characterizing the carbon nanotexture, ideally before and after neutron irradiation, can thus provide valuable insight into the alteration process and likely thermomecanical behaviour of the fuel particles.

Experimental

Specific software tools were developed for this study, designed from the start with digital imaging in mind. Since we wished the software to be easy to maintain, modify and transfer to other users, we opted for the Python language and its interactive development environment IPython [5, 6]. Python is a popular, multi-platform high-level programming language and possesses easy-to-use and powerful scientific libraries (*NumPy*, *SciPy* and *Matplotlib* [7, 8]). We also made use of the *Python Imaging Library* (Secret Labs AB, Sweden). However, several computer-intensive routines were (re-)written in ANSI C for performance purposes.

The procedure works as follows.

First, a 2D Fourier transform (FT) is performed on the LF image (Fig. 2). The power spectrum thus obtained is remapped onto polar coordinates, so that radial and/or angular intensity profiles may be extracted easily.

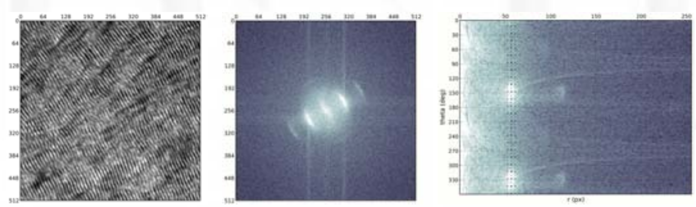

Figure 2: First steps of LF image analysis: Fourier transform (middle) and polar transform of FT (right).

Then, the FT radial profile (averaged within an estimate of the span of the 002 arcs) is fitted using an *ad hoc* model (Fig. 3). This yields the 002 ring position (r_{002}) and width (δr, Half Width at Half Maximum). From these values, one can obtain the average inter-layer spacing $\bar{d}_{002\ LF} = 1/r_{002}$, as well as the associated parameter $\Delta d_{LF} = 2\delta r/r_{002}^2$.

Next, the angular profile of the 002 ring is fitted using a gaussian-based model adjusted for 360° periodicity (Fig. 3), as previously described for diffraction pattern analysis [1]. This provides the local Opening Angle value (OA_{local}), which is a measure of the degree of mutual twist disorientation of the graphene layers.

We subsequently perform a spatial frequency (bandpass) and directional filtering of the original LF image, so that only 002 fringe features are retained (Fig. 4).

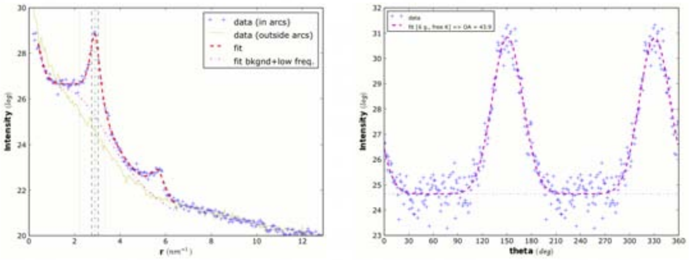

Figure 3: Fits of FT radial (left) and angular (right) intensity profiles.

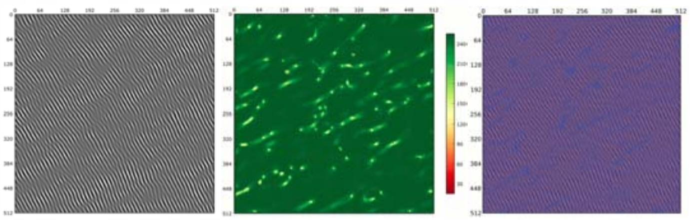

Figure 4: Extraction of lattice fringes primitives (right) from the filtered image (left); middle: confidence image.

The image then is further processed using pattern analysis functions [9, 10]. First is computed a "confidence" map, i.e., a measure of the reliability of fringe orientation estimation (Fig. 4). As areas with lower confidence values coincide with fringe endings, they are excluded from fringe detection. Image analysis proceeds with fringe extraction (Fig. 4), using a level curve tracking algorithm. This yields the length (L_2) and tortuosity (τ) of every detected fringe (Fig. 5).

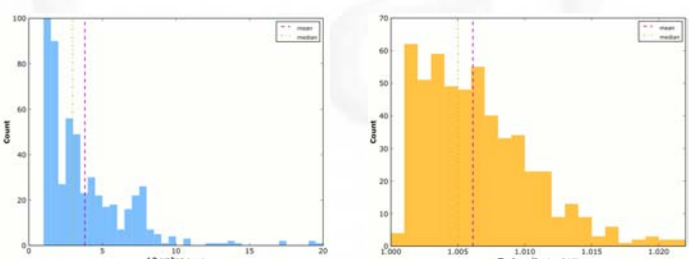

Figure 5: Distributions of the L_2 (left) and τ (right) parameter values in the LF image.

In addition, the confidence image can be used to estimate the density of nanotexture defects. We define the ratio ρ as the relative area whose confidence value is below the chosen threshold. ρ increases with the amount of defects in the field of view.

Results

We applied the methodology described above to the analysis of LF image series from several TRISO fuel particle productions.

Overall, we find much lower Opening Angle values than in SAED [1], as, at this scale, the OA measurement is mostly sensitive to local disorientations within the roughly parallel graphene layers, rather than to the material's overall curvature. Besides, the evolution of OA_{local} with the sample thermal history mirror what is observed in SAED.

The variations of the structural parameters $\bar{d}_{002\ LF}$ and Δd_{LF} are very similar and reflect the thermal maturation of the pyrocarbon. The two parameters are however not quite interchangeable.

Logically, we observe an increase of the average 002 fringe length and a decrease of the mean tortuosity (as well as of OA_{local}) with successive heat tratments, consistently with the increase in maturity shown by d_{002}. The variations of the density of defects ρ closely parallel those of τ, as it decreases with increasing apparent maturity in much the same fashion.

In addition, despite similar synthesis conditions, we were able to show significant differences in behaviour between sample series, apparently consistent with their failure rate under irradiation.

Conclusion

We have developed a methodology and the corresponding software tools for extracting quantitative structural ($\bar{d}_{002\ LF}$, Δd_{LF}) and textural (OA_{local}, L_2, τ, ρ) information, at the nanometer scale, from high resolution TEM images of carbon materials. It has been validated on dense pyrolytic carbon deposits from TRISO nuclear fuel particles and shown to provide relevant and valuable information on the samples' nanotexture.

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