Materials Characterization by Digital Microscopy

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Abstract

Materials characterization and analysis is the central theme of materials science. While computer-based methods greatly extend the scope of characterization techniques, these methods are associated with their own set of implementation issues in terms of image processing, statistical, and mathematical problems. Classical metallography does not prepare students for these challenges. We have developed a graduate course on microstructural characterization, analysis, and modeling that is based on concepts of stochastic microstructures and uses model systems based on spatial geometry concepts of point processes, packings, and tessellations. Using discrete constituents such as discs, we develop the fundamental ideas of spatial geometry and image algebra more transparently to aid student comprehension. Once these principles are covered, we extend them to more complex structures such as multiphase materials.

Introduction

This paper discusses our experiences with a 3-semester-hour (2 lecture and 2 laboratory hours) course taught to graduate students in the Department of Mechanical Engineering at NC A&T State University; these students were from different research groups and facilities but worked within the framework of the NSF Center for Advanced Materials and Smart Structures (CAMSS) for this special topic course. Their thesis topics ranged from computational mechanics to traditional materials science topics (semiconduction, nanomaterials), the materials primarily being composites (carbon/polymer, carbon/carbon, ceramic/ceramic) or monolithic ceramics. The topic of the course was the evolved equivalent of “Quantitative Metallography”, the examination of microstructures in the optical and scanning microscope ranges.

Because of the students varied background paths to the program, it was necessary to supply collateral support in statistics, spatial geometry, computer topics, and the property-structure link. The presentation of sufficient amounts of these topics in an integrated manner provided the major challenge for the course.

Course Scope

Like other areas, materials science is evolving a new relationship to the computer, which is bringing a strong link with mathematics and statistics. This is raising the expectations of microstructural analysis; however, an interpretative and analytic strategy is still needed and the translation of microstructure into the framework of computation or model is rarely trivial. This has resulted in an increased scope for the typical microstructures course, transforming it from metallurgy to materials.
A precis of the course topics follows.

1. Properties, Structure, Mechanisms
   1.1 Property-structure link
   1.2 Structure, geometry, images
2. Structural Geometry
   2.1 Packings, point fields, tessellations
   2.2 Parameters of single objects
   2.3 Parameters of arrangements of objects
3. Image Processing
   3.1 Image algebra
   3.2 Feature detection and extraction
4. Spatial Statistics
   4.1 Stochastic models
   4.2 Statistical Estimators
   4.3 Anisotropy, texture, orientation
5. Complex Microstructures
   5.1 Dispersoids
   5.2 Alloys

The topics were apportioned between lecture and laboratory; each student also had a major project focusing on their current or prospective thesis material.

Course Issues and Strategies

Because microstructural characterization and analysis is such a collective topic, involving other specializations, it presents difficulties in integrating these topics. We have found that using fibrous composites as our leading case has many advantages. A fiber is a cylinder in $\mathbb{R}^3$ Euclidian space which appears as an elliptic disc in the $\mathbb{R}^2$ Euclidian image plane, a group of these forming a packing; they can be further converted to a center of mass point field. From the packing point of view the statistical concepts of variation, and distributions, in shape and size, conveniently obtain. Consideration of the volume fraction also allows introduction of the idea of arrangement effects; we show the classic $\mathbb{R}^2$ packing arrangements by placing and analyzing coins on a scanner in hexagonal, cubic, and random arrangements. The point field allows introduction of such topics as nearest-neighbor, $K$-, and radial distribution functions. These are important for revealing interactions and correlations among the data. Tests using Poisson distribution are used to introduce complete spatial randomness. See [1] for a standard reference for these topics.

Voronoi and Delaunay tessellations are formed on the point field to partition the space; these are a standard way to analyze random structures. They are also a natural way to approach the much more complex topic of grains. See [2] for the standard reference on these topics.

The course used Image-Pro 4.5 [3], commercial image processing software that has a number of functionalities suitable for materials research, along with good statistical support and graphics output. However, the ease of obtention of some parameters has not changed the limitations of stereology, the $\mathbb{R}^3$ to $\mathbb{R}^2$ problem. It appears to us that the relationship between the image and the specimen space requires considerable effort to be appreciated. Unbiased stereology (not requiring...
any assumptions about shape or arrangement) is difficult in materials science, although it is required in medical research in many cases. This means that the various classical stereological parameters still prevail and must be clearly understood.

Examples

We present several examples taken from student projects, which illustrate our approach.

1). SiC/SiC composites

SiC/SiC composites exhibit excellent tensile (high toughness, strength and strain) and fatigue characteristics at ambient and elevated temperatures. The primary challenge in the commercialization of these composites has been to come up with an economical manufacturing process. Some of the current processes such as Chemical Vapor infiltration (CVI) suffer from the drawback of extremely long processing times combined with need for high processing temperatures and intermediate machining to decrease the residual porosity. The melt-infiltration (MI) process on the other hand is a low cost processing technique for the production of silicon based ceramics and composite materials. This process consists of the production of a microporous carbon preform and its subsequent infiltration with liquid silicon or refractory metal-silicon alloys. The microporous preforms are made by the pyrolysis of a polymerized resin mixture with very good control of pore volume and pore size. The process has near-net shape capabilities and complex shapes can be fabricated. The process gives very good control of the microstructure of the resultant materials.

The goal of the project was to compare microstructure of four different melt infiltrated SiC/SiC composites: 8-harness melt infiltrated Hi-Nicalon™ SiC fiber in SiC matrix ceramic composite (I), 5-harness Hi-Nicalon SiC fiber in SiC matrix ceramic composite (II) and a 5-harness Hi-Nicalon SiC fiber in a Si-rich SiC matrix composite (III) produced by NASA and 5-harness Hi-Nicalon SiC fiber in a Si-rich SiC matrix composite (IV) produced by Honeywell (former AlliedSignal Composites Inc.).

Figure 1 shows cross-sectional view of these materials indicating different porosity size and distribution in materials (I-IV). Materials I and II have mostly large voids, material IV has many small voids (pores). It can be seen that melt infiltrated Si-rich areas (bright spots) have different distribution in materials I-III and material IV has substantially different matrix material (gray). Within a tow there are smaller pores indicating places between fibers unreachable for infiltration (Figure 2, see Pores within Tow, Table 1). Also it can be seen that diameter of fibers varies considerably. Different materials have a different overcoat thickness (Figure 3). All of these features were analyzed quantitatively using extensive statistical analysis. Some of these results are presented in the Table 1. This is an example of the data routinely determined via image processing software. Detailed analysis of the data in comparison with mechanical properties of these materials exceeds the framework of this paper and will be presented separately.
Figure 1. SEM micrographs of polished samples of materials I-IV.

Figure 2. Optical micrograph of tow for composite II (magnification 200X).

Figure 3. Fiber overcoat structure in composites I-IV
Table 1. Results of image analysis of composites I-IV.

<table>
<thead>
<tr>
<th>Material System</th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean Fiber Diameter (µm)</td>
<td>12.41</td>
<td>13.05</td>
<td>13.48</td>
<td>12.85</td>
</tr>
<tr>
<td>Overcoat thickness (µm)</td>
<td>5.66</td>
<td>11.46</td>
<td>19.54</td>
<td>4.44</td>
</tr>
<tr>
<td>Number Fibers in Tow</td>
<td>525</td>
<td>590</td>
<td>563</td>
<td>491</td>
</tr>
<tr>
<td>Mean Fiber Density in Tow (µm²)</td>
<td>0.00331</td>
<td>0.00366</td>
<td>0.00702</td>
<td>0.00304</td>
</tr>
<tr>
<td>Pores within Tow (%)*</td>
<td>10.3</td>
<td>6.8</td>
<td>9.3</td>
<td>4.5</td>
</tr>
<tr>
<td>Grouped Large Voids (%)</td>
<td>7.0</td>
<td>6.9</td>
<td>3.7</td>
<td>3.5</td>
</tr>
<tr>
<td>Mean Void Area (µm²)</td>
<td>934</td>
<td>1954</td>
<td>680</td>
<td>426</td>
</tr>
<tr>
<td>Grouped Small Pores (%)</td>
<td>1.47</td>
<td>0.54</td>
<td>1.58</td>
<td>1.13</td>
</tr>
<tr>
<td>Mean Pore Area (µm²)</td>
<td>25.1</td>
<td>24.6</td>
<td>22.2</td>
<td>26.5</td>
</tr>
<tr>
<td>Si-Rich Area (%)**</td>
<td>3.62</td>
<td>4.57</td>
<td>8.54</td>
<td>30.45</td>
</tr>
</tbody>
</table>

Note: * – values calculated separately from pores for whole sample
**– represents area of matrix in composite IV.


Carbon-carbon composites are an exceptional class of high-temperature materials that have low density and very high temperature capability. The purpose of the project was to study the microstructures of a carbon/carbon composite during its different processing stages towards carbonization. The primarily focus is on the composite while it is post-cured, capturing cross-section images to analyze fiber volume fraction, which is an important parameter affecting the composite’s mechanical properties. The fiber density (number/area) in tow and unit cell, fiber radial distribution and average fiber diameter etc. are examined, comparing voids and cracks’ change after different manufacturing steps.

The composites were produced at the Center for Composite Materials Research (CCMR) at NC A&T State University. Fibers were Amoco’s T300 balanced 8-harness satin weave with a 3K size tow, equal number of yarn count in x and y directions, heat-treated under 2500 °C, matrix – Lonza’s Primaset PT30 Cyanate Ester Resin (thermoset) with a char yield of 65%, Fabrics System – Resin Transfer Molding of T300/PT30 Composite Flat Panels.

Three specimens were studied by the student from this center: 1) post cured; 2) following 3 carbonization/densification cycles, and 3) graphitized at 2500 °C. Analysis of samples from different stages of manufacturing process shows that voids and cracks which were created during curing stage are almost filled out after 3 carbonizations, then appear again after graphitization (Figure 4). Average distance of voids and cracks are 292.37, 268.63, and 210.11 µm respectively. Void and crack directions are normally perpendicular to the fiber direction and from surface to surface across the entire tow.

Figure 5 shows a tow-based representative structural element. High-resolution images (1600x1200 pixels) allow performing accurate counting and packing analysis of fibers within tow at relatively low magnification (200X).
Figure 4. Changes in crack system during carbon-carbon composite manufacturing procedure (magnification 100X).

Figure 5. Carbon/Carbon composite tow analysis: a) unit cell with 2 tows (combined from 6 images at magnification 100X); b) tows (200X): fiber volume fraction in unit cell = 22.38%; fiber density in unit cell = 0.00923 μm^2; fiber density in upper tow = 0.0179 μm^2; fiber density in lower tow = 0.0178 μm^2; ratio for fiber density in Tow/Unit cell = 1.93; average number of fibers in one tow = (2991+3010)/2 = 3001 (3K expected).

Fiber diameter distribution, packing number and the radial distribution have been obtained at magnification 1000X (Figure 6). It shows that fibers have non-cylindrical shape and surprisingly sharp distribution with a mean diameter of 6.35 microns (SD=0.21 for 170 samples). Average contact number is 3.54, without deep valleys between radial distribution peaks and twice the fiber density in first peak in radial distribution compare to average fiber density indicate medium order between fibers in tow. A fully ordered system (cubic or triangular) has an ideally expected average spacing between fibers of 7.58 microns; a Poisson distribution has an 3.79 micron expected spacing. Therefore, the experimentally obtained value 6.43 micron is towards the ordered end of values. It should be mentioned that average fiber densities obtained at high magnification were the same as obtained from tow analysis at 200X.
3). Image analysis of porous materials

Analysis of porous materials is complicated because it requires \( \mathbb{R}^2 \) reconstruction of image from \( \mathbb{R}^2 \) images. Scanning electron microscopy (SEM) has wide depth of field and gives sharp images from porous surfaces, but quantitative \( \mathbb{R}^3 \) reconstruction from these images is extremely difficult. High-resolution traditional optical microscopy has a sharp focal plane which does not allow obtaining the crisp images of non-flat surfaces but it can generate a \( \mathbb{R}^3 \) topographical map of the surface using an extended depth of field technique.

One of the student projects was concerned with comparison of porosity in different samples of MgB\(_2\). Technical applications of this material require as much densification as possible. Analysis of the porosity level can be performed if we can separate voids and material from \( \mathbb{R}^2 \) image; SEM images are not useful for this. Optical images of rough surface of particles are sharp in the focal plane at high magnification therefore we need to obtain multiple images. The extended depth of field technique allows us to combine a sequence of collected at different focal distances images into a single, in-focus composite image. Figure 7 shows extended depth of field image reconstructed from 20 images and the topographical map showing 20 different focal planes. It can be seen that the surface profile reproduces bright areas in the extended depth of field image relatively well. Using this approach the value 46.2\% of porosity within MgB\(_2\) particle was obtained.

We can conclude that this approach uses the apparent limitation of a sharp focal plane for the optical microscope to establish topology. Accurate analysis of sequences of images makes the optical microscope more valuable here than a SEM.
Figure 7. Extended depth of field image of MgB$_2$ particle (left) and its topographical map.

Summary Observations

These examples typify contemporary capability of imaging software. It requires considerable time for students to learn sufficient details about the software to make good use of the wider array of image processing and analysis features that might be commonly used. It is also inevitable that macros need to be written for custom combinations of operations. We found that considerable lab instruction and practice time was needed for this, and that a single license was insufficient for a class of nine students.

The aforementioned ease of obtaining microstructural parameters accents the need to understand the limitations of stereological equations, and their probabilistic basis. While we did not prove all of the classic equations, we argued their form and obtained bounds using cylinder, sphere, or cube packing geometries. The multitude of available parameter statistics also required a review of elementary statistics and distributions.

It is also necessary to stress that without auxiliary information, such as microhardness traverses, electrical properties, knowledge of the components or phases, or process or forces to which the structure has been subjected, microstructural examination is pointless. This means that the students need to bring to, or acquire in, this information for the course. We found it necessary to devote some time to mechanisms which might be operative in a particular microstructure. An example is a Weibullian flaw, formed by the complex of clustered fibers and weak interfaces. We stressed that an interpretative strategy was necessary for microstructural examination, and that even selection of a feature to be examined is a form of modeling.
References

3. Image-Pro Plus, Media Cybernetics Silver Spring MD 20910

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