THREE DIMENSIONAL MICROSTRUCTURE OF POLYMERIC COMPOSITE MATERIALS USED IN SANDWICH STRUCTURES USING DUAL MODALITY FROM COMBINED HIGH RESOLUTION X-RAY AND NEUTRON TOMOGRAPHY

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Abstract
Use of dual modality to study polymeric composite materials with high resolution X-ray and neutron tomography is demonstrated for the first time in this study. X-ray and neutron tomography techniques provide an ability to visualize and quantitatively describe the microstructure in three dimensions (3-D) non-invasively. 3-D image based registration is performed to combine the two modalities that account for varied resolutions and contrast. Authors are studying the effect of degradation of carbon fiber vinyl ester based composites due to sea water exposure and the damage diagnostic techniques from tomography, and are finding beneficial use to understand the complex coupling of microstructure-mechanical property relationships. Quantitative information such as local versus global variation of fiber and resin volume fraction, analysis of voids or cracks, and anisotropy of attenuation properties are being evaluated in 3-D for use in studying these complex materials used for marine ship sandwich structures.

1 Introduction

Carbon fiber reinforced polymer composite materials are used as structural materials in the construction, automotive, and aerospace industries for their high specific strength and stiffness, corrosion and fatigue resistance and ease of handling and fabrication [1]. Carbon fiber composite materials are used as the facing materials for sandwich structures, and sandwich structures have been investigated for possible use in naval application. The environmental degradation due to long term exposure of sea water effects on the composite facing material is a significant research problem which is actively being evaluated [2, 3].The observed degradation and related mathematical mechanics based modeling of constitutive material properties of facing, foam core, and interfacial fracture of sandwich structures due to harsh sea environment is currently based on the observation of macroscopic laboratory data, and authors are evaluating micro-tomography data to further enhance the related basic research. Radiation (X-ray and neutron) based imaging can provide important information about the state of microstructure of the composite sample in terms of the distribution of various phases corresponding to fibers, resin, manufacturing defects, and the underlying damage and failure mechanisms when coupled with in situ mechanical testing systems. Coupling of powerful tomography based imaging systems with proven and well calibrated ultrasound based diagnostic techniques have significant future potential in such studies.

In order to visualize the interior of a sample, imaging technology has been applied in the present paper to study carbon fiber and vinyl ester based polymeric composite facing materials. Until recently, microstructure for composite materials is obtained by destructive techniques such as thin sectioning of impregnated and damaged samples, subsequent careful polishing, obtaining two dimensional images from optimal microscopy followed by digital image analysis. This method is not only time consuming, but also prone to have errors such as non-parallel sectioning, fiber shape distortion during the sample preparation, and mismatch of fiber between adjacent dissected surfaces [4]. X-ray tomography method has been used as an alternative. Lab based microtomography (micro-CT) machines are readily available and provides resolutions in the order of a micrometer or less. The highest resolution is achievable at

synchrotron imaging facilities [5] using monochromatic X-rays. X-ray tomography techniques have been applied to study damage from mechanical and thermal stresses and UV radiation for polymeric composites [6-8]. Synchrotron tomography imaging experiments coupled with conceptual in-situ mechanical tests on carbon fiber laminated epoxy composite materials were recently reported with impressive spatial resolution of visualizing individual fibers for damage analysis and shows promise [9-12].

For the first time, authors performed simultaneous X-ray and neutron tomography experiments on wet and dry carbon fiber polymeric composite specimens using unique imaging resources at Helmholtz Center Berlin for Materials and Energy (HZB), Berlin, Germany. The objective of this paper is to present a new concept of studying polymeric composite materials using combined use of X-ray and neutron tomography techniques to exploit the different contrast possible from two types of radiation. The contrast of neutron and X-ray tomography data is compared, and spatial registration of sample in 3-D using the two modalities is also demonstrated. The methods developed in the research will be extended to future in-situ loading experiments to study the failure mechanism of wet and dry carbon fiber composite materials to understand the fundamental stress-strain behavior and damage evolution under extreme environments.

2 Sample Description and Experimental Procedure

2.1 Sample Preparation

The carbon fiber composite material in this study was made of carbon stitch bonded fabric designated as LT650-C10-R2VE supplied by the Devold AMT AS, Sweden. This was an equibiaxial fabric produced using Toray's T700 12k carbon fiber tow with a vinyl ester compatible sizing. The individual carbon fiber diameter is 7 μm. The resin matrix used was Dow Chemical's DERAKANE 510A-40, a brominated vinyl ester, formulated for the Vacuum Assisted Resin Transfer Molding (VARTM) process. The bromination imparts a fire-resistant property to the composite. Carbon fiber reinforced/vinyl ester laminated composites consisting of $[90/0]_{2s}$ cross-stitched lay-ups are used in this study. Test materials were fabricated following established procedures [13]. Coupon samples obtained from the face sheets were then used for this study. One batch of samples was kept dry (allowed to age) and another batch was soaked by immersing the samples in a bath of sea water at 40 ˚C for at least 6 months prior to testing. This duration was sufficient to reach equilibrium sea water uptake based on periodic weight gain measurements. Generally the equilibrium state for sea water diffusion was reached upon immersion within approximately three months, with a weight gain of about 0.4%. Two samples with the dimension of 25.4 mm × 50.8 mm × 2.8 mm from large sheets were prepared by getting a coarse sample using a diamond saw and sample with final dimensions using surface grinder with diamond blade. These dry and aged sample and wet (soaked) sample were transported to imaging facility in sealed bags for tomography experiments.

2.2 Neutron Tomography and X-ray Tomography

X-ray tomography was performed using a laboratory microfocus X-ray machine. Neutron tomography was performed at a dedicated neutron imaging beam line at HZB called the cold neutron radiography and tomography (CONRAD) with cold neutron spectrum coming from a 10 MW reactor using the high resolution setup with CCD camera and a 10 μm thick gadox scintillator [14, 15]. The imaging parameters are provided in Table. 1. Both systems provided relatively large field of view (FOV) and spatial resolution adequate for engineering applications.

High spatial resolution is required to visualize the fine features of the sample. However, increasing the spatial resolution inevitably reduces the achievable FOV in general. The neutron imaging detector used in this research can achieve up to 13.5 μm/pixel resolutions with 27.6 mm × 27.6 mm FOV. The FOV and spatial resolution can be changed by varying the working distance and magnification of the lens. Even the highest resolution (13.5 μm/pixel) would still be too coarse to visualize the individual fiber (7 μm dia.). As a result, the FOV was adjusted to visualize the entire sample area. In case of microfocus X-ray tomography system, the magnification was adjusted to visualize the top half of the sample with 13.2 μm/pixel resolution. For many engineering applications and simulations, it is

necessary to choose the appropriate FOV and spatial resolution for the length scale of interest. The result from the image data can be compared to the simulation data at the similar length scale as the length scale affects the simulation result.

2.3 Computed Tomography Reconstruction

When neutron or X-ray beams pass through a sample, some of them are attenuated (scattered or absorbed). The amount of attenuation is characterized by the total macroscopic cross section (Σ_T) for neutron or the attenuation coefficient (μ) for X-ray. The attenuation process is linear and can be presented as the Lambert-Beer's law for monochromatic beam as in eq. (1) where I_0 is the incident beam intensity and I is the beam intensity after passing the material.

$$I = I_0 e^{-\int \mu(x)dx} \qquad (1)$$

Computed tomography experiment is performed by taking several projections at different angular positions of the sample. All projections can be put together to reconstructed a tomography slice which shows the spatial distribution of the attenuation value in the interior of the sample. The reconstruction is commonly done with an algorithm such as filtered backprojection as well as iterative algorithms [16]. The given samples were rotated 360° with 400 projections for neutron tomography and also 360° rotation with 1000 projections for X-ray tomography. They were reconstructed by using the filtered backprojection algorithm of Octopus 8.4

2.4 Multimodality of X-ray and Neutron

The interaction mechanisms of neutrons and X-rays with a material are different. Neutrons interact directly with the nucleus of an atom while X-rays interact with the electron cloud of an atom. As a result, X-rays' cross sections (attenuation) increase with the atomic number while neutrons don't have a particular trend. For example, X-rays have a low cross section for hydrogen and hydrogen containing material, but have a high cross section for metals like steel and aluminum. On the other hand, neutrons have a high cross section for hydrogen, but have a low cross section for steel and aluminum. Such different attenuation behavior can be advantageous to study some mix of materials. The multimodality of neutron and X-ray tomography on partially water saturated silica sand sample placed in an aluminum sample holder is shown in Fig. 1 [15]. The partially saturated silica sand has silica (SiO_2), water (H_2O), and air phases. It is easy to identify the water phase from neutron data, and silica phase from X-ray data. Such multimodal contrast has good potential for imaging composite materials such as glass fiber reinforced composite materials.

3 Results and Discussions

3.1 Registration

A 3-D image registration method was used to combine the multimodal volumetric data. The registration techniques have been popularly used in medical imaging to combine multimodal images for easier diagnosis and surgical planning [17, 18]. Similarly, X-ray tomography images and SEM images of a soil sample has been registered [19]. Multimodal registration of neutron and X-ray data was also tried [20]. The two volumes are compared and registered based on correlation method due to the similar contrast mechanism [18]. The registration is performed based on the image intensity and doesn't require markers. Rigid body rotations and isotropic scaling was performed to find the best orientation of registration. The registration process was performed with the affine registration module of Avizo 6.3. Orthoslices of X-ray and neutron data at the same location after registration is compared in Fig. 2. The idea can be extended to registration of variable size of FOV/resolution data together.

3.2 Image Processing and Phase Quantification

Approximately 10.5 mm × 13.2 mm × 2.1 mm sub-volume of X-ray data of the dry specimen was chosen for the analysis. The 3-D microstructure of dry specimen is shown in Fig. 3. The resin has a higher attenuation than the fibers. In order to segment the resin phase from the fiber phase, the factorization thresholding method by maximizing the interclass variance was applied [21]. The thresholding result is shown in Fig. 4. The resin volume fraction was found to be approximately 15.6 % based on thresholded results.

3.3 Resin Distribution

As shown in Fig. 3, the resin of horizontal direction had lower density compared to that of vertical

direction. X-ray tomography provides means to characterize and quantify the resin distribution. Such directional difference with the resin density can affect the mechanical strength of the composite material.

3.4 Sea Water Effect

Recent research of Drs. Penumadu and Siriruk show that the cyclic fatigue behavior of wet and dry carbon fiber reinforced composite samples ($[\pm 45]_{2s}$) varied dramatically with the number of cycles to failure decreasing substantially for wet and water confined composite samples [22]. X-ray micro-CT shown in Fig. 5 revealed important fundamental damage evolution process different for dry and air confined when compared to wet and water confined corresponding to immersed conditions. It was found that the fatigue life was reduced up to 85% when cyclic loading was applied under the immersed condition compared to the dry condition. Fig. 5 shows that the main mode of failure for wet and confined samples is delamination of outer layer, where as for dry sample the entire sample took part in providing mechanical resistance during cyclic loading.

3.5 Beam Hardening Artifact

Lab based micro-CT machine and reactor based neutron tomography beam line generally use polychromatic beam. The attenuation is energy dependent, and the lower energy neutrons or X-rays have lower penetrability than the higher energy neutrons or X-rays. The difference of penetrability occurs more significantly when the beam is passing through a longer path length. In Fig. 6, more significant attenuation occurred when beam penetrated through the width of the sample than through the thickness since much more lower energy X-rays didn't get detected. Such beam hardening shows up as cupping artifacts in which the reconstructed attenuation value decreases toward the center of the object in the reconstructed slices. Fig. 7 shows that the reconstructed attenuation value decreases toward the middle of the sample almost 40 % in case of X-ray tomography data. Similar artifact appeared on neutron tomography data as well. Beam hardening artifact makes it difficult to perform image segmentation and quantification.

4 Conclusion

Simultaneous use of high resolution X-ray and neutron tomography was introduced for studying polymeric composite materials and sandwich structures. Use of dual modality (contrast from X-rays and neutrons) obtained from imaging systems that have different spatial resolution and detector systems needs a combined 3-D registration scheme for visualization and quantitative analysis. In this paper, registration technique was introduced as a method to combine multimodal and variable FOV/resolution data. Example use of proposed non-destructive imaging techniques to study sea water induced degradation in mechanical properties of carbon fiber vinyl ester system is included. The high resolution imaging capabilities are being extended to in-situ testing of composite materials for damage visualization and quantification at different loading stages, and are parts of ongoing research.

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Instrument	Peak Energy (keV)	Voxel Size (μm)	Detector Pixel Size	FOV (mm)
X-ray	100	13.2	2316 × 2316	30.5 × 30.5
Neutron	7.30E-06	29.8	2048 × 2048	61 × 61

Table.1. Imaging parameters of X-ray and neutron imaging systems

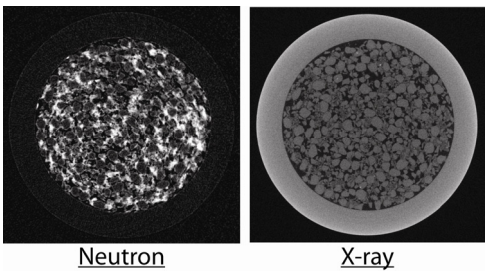

Fig. 1: Neutron and X-ray multimodal contrast comparison of partially saturated sand sample composed of silica, water and air phases

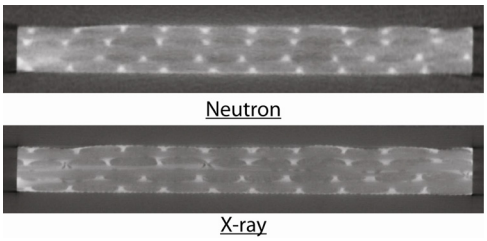

Fig.2. Comparison of neutron and X-ray orthoslices at the same location

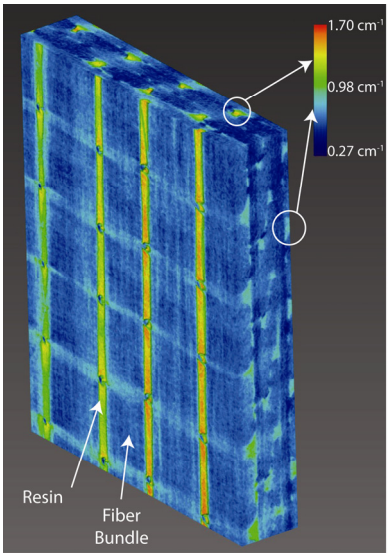

Fig.3. 3-D view of the sub-volume with a color map of attenuation coefficient

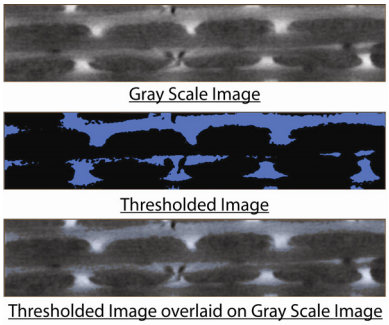

Fig.4. Thresholding result

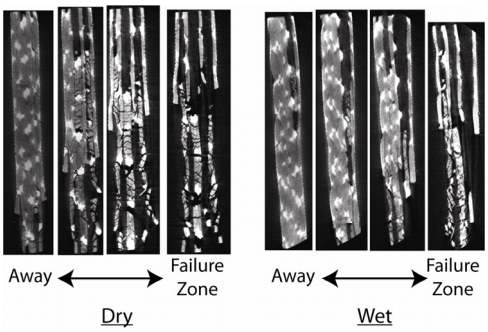

Fig.5. X-ray tomography slices of $[\pm 45]_{2s}$ dry and wet specimens loaded to failure during cyclic loading

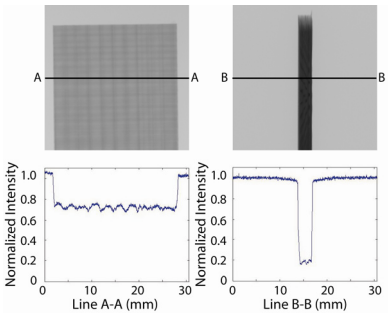

Fig.6. X-ray radiography profile comparison

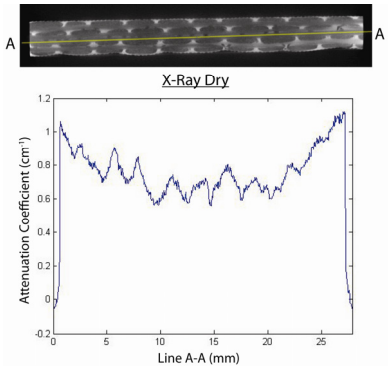

Fig.7. Profile of X-ray tomography slice showing beam hardening artifact

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