Investigation of modern engineering materials using laboratory small angle X-ray scattering

Susanne BURTZLAFF 1, Stefanie HÜBNER 2

1 Project group NanoCT Systems, Fraunhofer Development Center for X-ray Technology EZRT; Wuerzburg, Germany; Phone: +49 931 3185497, Fax: +49 931 3181909; e-mail: susanne.burtzlaff@iis.fraunhofer.de
2 Application-specific Methods and Systems, Fraunhofer Development Center for X-ray Technology EZRT; Fuerth, Germany; e-mail: stefanie.huebner@iis.fraunhofer.de

Abstract
Particular requirements are imposed on modern composite materials used in aerospace. Thus special care has to be taken to test the stability and endurance of these materials. An alternative approach will be presented to investigate the properties of the composite materials in a mesoscopic scale. Small Angle X-ray Scattering (SAXS) is caused by elastic scattering of X-ray photons on mesoscopic particles and interfaces. The angular scattering signal contains information about the average size, shape, specific surface and orientation of the particles. The principle of the SAXS technique will be presented along with practical applications on fiber composite materials. The spatially resolving measurement of composite materials will reveal the general structure composition along with local disorders.

Keywords: Other Methods, Radiographic Testing (RT), composite, scattering, X-ray, SAXS

1. Introduction
Several methods are complementary used in material characterization and nondestructive Testing to describe the various aspects of the investigated material. Defects like delamination are challenging most of the conventional methods. X-ray radioscopy and computed tomography are used to determine the transmission. Differences of the density, the atomic number or the thickness of the material are causing contrast and hence allow the determination the structure or defects of the material. These methods are often combined with thermography [1]. The heat transfer is specific for any material and analyzing it indicates possible disturbances inside the material. Hentschel et al. presented a refraction based method which basically roots in the theory for Small Angle X-ray Scattering [2]. Using this method he is able to determine orientation distributions and inner surface densities. SAXS is a technique which is based on elastic scattering of X-ray photons in materials. At interfaces the scattering is directed specifically for the scattering contrast and the angle of the interface [3]. As an interface sensitive technique it is suitable to analyze delamination e.g. for fiber matrix systems.

2. Investigation of CFRP using Small Angle X-ray Scattering
For this investigation a 150 x150 x 2 mm³ plate of carbon fiber reinforced plastic CFRP was used. The fiber structure is single layered with a vertical orientation. The impact was placed on the back site of the CFRP. The back site appears to be intact beside a diagonal mark of the impact with a length of about 15 mm. On the front side the structure is destroyed at the place of impact and partly ripped out.

Beforehand an X-ray radiography was taken (see figure 1). It revealed that additionally to the carbon weave some glass fibers have been inserted; these are visible due to their higher absorption. The crack itself is barely visible. Other damages than that cannot be determined.
The thermography analysis reveals the damage much better but it is still hard to determine how far reaching the damage really is.

The reconstructed slice of an X-ray computed tomography measurement reveals a fine crack along the carbon weave. However the possibility to identify the range of such delamination is limited by the resolution of the reconstructed volume.

The force of the impact is transported by the fibers. The impact destroys bigger particles. So in the damaged volume there have to be more small particles than in the intact volume. An accumulation of smaller particles indicates a higher damage.

One possibility to get statistical information about the average size of particles in a limited fraction of the complete volume is the Small Angle X-ray Scattering SAXS.
2.1 Setup and Data acquisition

The experiments have been performed on a self-developed system.

4: Sketch of the measurement setup.

In figure 4 the setup is schematically presented. The X-ray beam will be shaped by the collimators. At the sample the beam will be scattered. In a certain distance the detector is recording the scattered beam.

5: Fully evacuated SAXS system with a) the rotating Cu X-ray source, b) the Göbel mirror, c) the collimation system, d) the sample stage and e) the detector tube with the MediPix2 detector on motorized axes.

To accomplish measurements on solid light materials such as CFRP a customized system has been developed to allow the analysis of big particles in the very small angle range and to enable scanning measurement.

The X-rays are generated using a rotating anode X-ray tube with a copper target. A Göbel mirror optic is parallelizing and monochromatizing the X-ray beam. Three movable blade slit systems are shaping the X-ray beam into a needle beam. This and other slit configurations are described by O. Kratky et al [4]. By passing through the sample the X-ray beam scatters in a material specific way. To detect the scattered X-ray beam the MediPix detector is being used - a sensor with small pixels, a very low noise and a high efficiency [5]. The high contrast between the prime beam signal and the scattering signal would normally cause a lot blurring. Due to the bumping-bond connection between the sensor and the readout of the MediPix detector, the internal blurring of the detector is low. That way a beam stop is not necessary and we are able to record the signal very close to the primary beam in a very low angle. By increasing the sample detector distance (ODD) up to 3 meters the observed scattering angle can be even lower. To prevent unwanted absorption and scattering by air, the whole system is evacuated. Little vibrations will cause smearing of the signal in that small scattering angle range. Hence the system is placed on massive granite.
The needle beam was shaped with a diameter of 300 µm to achieve an appropriate photon flux. To get a good angle resolution a sample detector distance (ODD) of 2923 mm was chosen. The sample has been scanned point wise on a distance of 3 to 73 mm from the place of impact.

2.2 Data treatment

The momentum transfer is expressed as $q$ as a wavelength ($\lambda$) independent angle unit which is inversely proportional to the particle size $d$ (see figure 6 and formula 1). The scattering angle $\theta$ has to be calculated using the distance to the beam center in pixels, the pixel pitch and the sample detector distance ODD.

\[ q = |\mathbf{q}| = 4 \pi / \lambda \sin \theta \]

6: Schematic illustration of the momentum transfer formation.

\[ d = \frac{2\pi}{q} \]

(1)

The measured image displayed an isotropic scattering. That means the structure of the sample did not have any preferential orientation. Due to that the azimuthal integration was performed across 360°.

7: Resulting scattering image.
The azimuthal profile is displayed in figure 8. The measurement without any sample \( (I_0 \text{ measurement}) \) produced a more narrow profile than the measurements with a sample. The measurement at the damaged part of the CFRP sample was performed in the middle of the sample where the impact was applied. The measurement at the part of the CFRP sample which was assumed to be intact was performed close to the border of the sample in a 70 mm distance to the place of impact. To get any clues about the different structures at these places the data had to be further processed.

8: Azimuthal profile of the scattering image with the empty beam measurement \( I_0(q) \) (red line) and the sample measurement \( I(q) \) (blue line).

By scaling the sample measurements \( I(q) \) by the transmission \( T \) and subtracting the \( I_0(q) \) measurement the scattering function becomes apparent.
9: Primary beam corrected scattering data.

In the low $q$ range the Guinier fit has been performed with the variables $G$ and $R_g$. The Guinier constant $G$ is defined by the differential scattering length density $\Delta \rho$ and the irradiated Volume $V_p$ (see formula 2). The radius of gyration $R_g$ is defined as the average distance of each position of the particle to the center of the particle $r_{\text{mean}}$ (see formula 3).

In the high $q$ range the Porod fit can be performed. The Porod constant is defined by the mean scattering length density $\langle \Delta \rho \rangle$ and the Surface per irradiated Volume $S_V$ (see formula 4).

\[
G = \Delta \rho^2 V_p^2
\]  
(2)

\[
R_g^2 = \frac{1}{N} \sum_{k=0}^{N} (r_k - r_{\text{mean}})^2
\]  
(3)

\[
PC = 2\pi \langle \Delta \rho \rangle^2 S_V
\]  
(4)

The CFRP has been scanned starting from the place of the impact damage up to the border of the sample.

In the profiles of the measurement points in increasing distance to the place of impact the high $q$ range slope is steadily dropping without any further features. The Porod constant does not change for the measurements of the CFRP along the scan. Due to that our analysis focuses on the high $q$ range and the Guinier analysis.
In figure 10 the radius of gyration $R_g$ has been plotted for all measurements. On the same scale a slice of a reconstructed volume of the CFRP sample is displayed.

![Graph showing the radius of gyration $R_g$ plotted versus the distance from the place of impact compared to a computed tomography volume slice of the composite plate (voxel size: 86.6269 µm)](image)

Figure 10 reveals a lot of inhomogeneities in the sample. Partly these are caused by the structure of the CFRP itself partly by the brought in damage. The radius of gyration is bigger in a distance of > 60 mm from the place of impact. That means that less particles have been destroyed there and this region can be assumed to be an intact region. The determined radii of gyration correspond to the results of the computed tomography. But due the statistical average over all the particles in the irradiated volume the contrast between the intact and damaged CFRP is only slightly. Nonetheless could the long range defects be identified. Whether the damage was caused by a broken matrix or by the fiber matrix delamination cannot be ascertained at a single point of measurement. But the extensive damage across a big distance from the place of impact indicates that the damage was transported along the fiber structure. Hence the damage in the far distance of the place of impact has to be caused by fiber matrix delamination.

### 2.2 Results

With SAXS a much more far reaching damage caused by the impact could be identified than it could be expected from the outer appearance of the sample, the radioscopic image (see figure 1) or the thermography analysis (see figure 2). The far reaching damage can be confirmed by the reconstructed volume of the sample (see figure 3). A fine vertically crack is noticeable up to a distance of about 50 mm from the place of impact.
The far reaching damage which was ascertained by computed tomography and by SAXS has to be caused by fiber matrix delamination. Using SAXS inhomogeneities are also very well detectable.

3. Conclusion

Small Angle X-ray Scattering is an appropriate method to detect hidden damages in materials provided they are causing a statistical relevant effect. This method has some limitations. Due to the small scattering angle soft X-ray has to be used. That is reducing the possible applications to light weight material investigation. For advanced analysis of the data some prior knowledge of the material is necessary. Laboratory systems also have to deal with low photon flux and hence long measurement times. The method can provide information about the sample which might not be accessible with other methods.

References