High resolution X-ray computed tomography of fibre- and particle-filled polymers

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Abstract
X-ray computed tomography (XCT) provides a volumetric map of a specimen in three dimensions, generated from a set of radiographs. Due to the speed and quality of measurement, XCT systems with cone beam geometry and matrix detectors have become widely used. Continuous improvements in the quality and performance of X-ray tubes and devices have led to cone beam XCT systems which can now achieve spatial resolutions down to 1 µm and even below. We report on the application of high-resolution X-ray computed tomography for the characterisation of various fibre-reinforced and particle fibre-reinforced polymeric materials. Due to their specific and outstanding properties for, for example, lightweight or packaging materials, they are widely used in the automotive, aeronautic, electronics, building, textile and leisure industries, as well as in medicine and agriculture. Fillers (particles and fibres) can be classified as inorganic, cellulose and polymeric fillers. We compare the different polymeric material systems and discuss the advantages and limitations of XCT for the characterisation of these systems. Due to varying density differences the contrast between the fibres and the polymeric matrix can be very good with, for example, glass- or talcum-reinforced polymers, or rather poor, as in the case of polymeric fillers within a polymeric matrix. Quantitative data can be extracted from the XCT-data. By applying various 3D-filters the following quantitative information and values can be extracted from the XCT-data:

- filler percentage
- 3D-geometry of the fillers (diameter, surface, volume)
- fibre length distribution
- fibre orientation (orientation tensor)
- 3D filler distribution
- filler interconnectivity

Keywords: X-ray computed tomography, phase contrast, polymers, reinforced polymers

1. Introduction
Fibre- and particle-reinforced polymers are important materials for many industrial branches like the aeronautics and automotive (e.g. for lightweight applications) and the packing, construction and leisure industries [1]. Fillers are used for reasons of economy and for reinforcement of the polymeric matrix. The properties of reinforced thermoplasts are mainly influenced by the percentage of the filler, its form (e.g. aspect ratio) and by the interaction of the filler with the matrix [1]. The most common fillers and materials are:

- inorganic fillers:
  - glass-fibre reinforced polymer
  - carbon-fibre reinforced polymer
  - basalt fibre reinforced polymer
  - talcum particle filled polymer
  - CaCO₃-particle filled polymer
  - wollastonite-filled polymer
  - mica-filled polymer
- cellulose fillers:
  - cellulose fibre filled polymer
  - wood plastic composites
  - cellulose particle filled polymeric foam
- polymeric fillers:
  - aramide fibre reinforced polymer
  - polypropylene blend containing rubber

In this publication we present the possibilities of using high resolution X-ray computed tomography (XCT) for the 3D-characterisation of fibre- and particle-filled polymers. X-ray computed systems with cone beam geometry have reached resolutions down to 1 µm [2]. For this reason and due to its many other advantages, CT has gained general acceptance in materials science for 3D-characterisation [2].

2. Experimental

2.1 Samples

A broad variety of different polymeric materials with different fillers were investigated. Table 1 shows a summary of the investigated samples together with the most important filler and XCT-measurement parameters.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Filler</th>
<th>Density of Filler</th>
<th>Form of Filler</th>
<th>Voltage</th>
<th>Voxel Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-polypropylene</td>
<td>Glass</td>
<td>2.6 g/cm³</td>
<td>Fibres</td>
<td>60</td>
<td>(3 µm)³</td>
</tr>
<tr>
<td>PA66-polyamide</td>
<td>Carbon</td>
<td>1.8 g/cm³</td>
<td>Fibres</td>
<td>60</td>
<td>(2 µm)³</td>
</tr>
<tr>
<td>Resin</td>
<td>Carbon</td>
<td>1.8 g/cm³</td>
<td>Fibres</td>
<td>60</td>
<td>(1 µm)³</td>
</tr>
<tr>
<td>Resin</td>
<td>Basalt</td>
<td>2.7-3. g/cm³</td>
<td>Fibres</td>
<td>80</td>
<td>(10 µm)³</td>
</tr>
<tr>
<td>PP-polypropylene</td>
<td>Talcum</td>
<td>2.6-2.8 g/cm³</td>
<td>Particles</td>
<td>40</td>
<td>(0.5 µm)³</td>
</tr>
<tr>
<td>PP-polypropylene</td>
<td>Mica</td>
<td>2.85 g/cm³</td>
<td>Particles</td>
<td>40</td>
<td>(5.5 µm)³</td>
</tr>
<tr>
<td>PP-polypropylene</td>
<td>4 % cellulose particles</td>
<td>1.5 g/cm³</td>
<td>Fibres</td>
<td>50</td>
<td>(0.7 µm)³</td>
</tr>
<tr>
<td>PP-polypropylene</td>
<td>Cellulose</td>
<td>1.5 g/cm³</td>
<td>Fibres</td>
<td>40-50</td>
<td>(2.5 – 5.25 µm)³</td>
</tr>
<tr>
<td>Wood plastic composite</td>
<td>Wood</td>
<td>Variable</td>
<td>Particles</td>
<td>85</td>
<td>(31 µm)³ and (5.5 µm)³</td>
</tr>
<tr>
<td>PU-polyurethane</td>
<td>Cellulose</td>
<td>1.5 g/cm³</td>
<td>Particles</td>
<td>50</td>
<td>(0.7 µm)³</td>
</tr>
</tbody>
</table>

2.2 High resolution cone beam XCT-device

Most X-ray tomograms were scanned using a “nanotom 180NF XCT” desktop device, developed and manufactured by GE Sensing & Inspection Technologies: phoenix|x-ray with a 180 keV high-power nano-focus tube with transmitting target and a 2300x2300 pixel Hamamatsu detector. Some of the XCT-measurements were performed using a Rayscan 250E XCT-device. The Rayscan 250E system is equipped with a 225 keV-µ-focus tube by Viscom and a 1024x1024 a-Si flat panel matrix-detector by Perkin Elmer. Targets made of
molybdenum were used at voltages between 50 and 85 kV. The voxel size used was between (0.4 µm)$^3$ and (31 µm)$^3$. These parameters required measurement periods of between 45 and 600 minutes. The X-ray tube of the “nanotom” is equipped with an external liquid cooling system to ensure stable measurement conditions and to minimise thermal influences during scans of longer duration. The cone-beam XCT data were reconstructed by a filtered-back projection Feldkamp algorithm [3]. The reconstructed data were processed and visualised with the programme VGStudio MAX 2.1. Further details can be found in [4] and [5].

3. Results and Discussion

3.1 XCT-results

Figure 1 shows a cross-sectional XCT-image and a three-dimensional visualisation of a short glass-fibre reinforced polymer. Due to the rather large density difference between the fibres and the polymer matrix, individual fibres are clearly visible in the XCT-data and can be segmented 3-dimensionally. The XCT-data can be used to extract fibre orientation and the fibre length distribution function [6].

Fig. 1. Cross-section of XCT-data three-dimensional representation of a glass fibre reinforced polypropylene sample (GFRP) produced by injection moulding. The voxel size was (3 µm)$^3$.

Fig. 2. Cross-section of XCT-data of a carbon-fibre reinforced polymer sample PA66 produced by injection moulding and a three-dimensional representation of a carbon-fibre reinforced polymer sample (CFRP). Voxel size was (2 µm)$^3$ for the left picture and (1 µm)$^3$ for the right picture.
The left picture of Figure 2 shows a cross-sectional XCT-picture and a three-dimensional visualisation of a short carbon-fibre reinforced polymeric part produced by injection moulding. Although the density difference between the carbon fibres (density ~1.8 g/cm$^3$) and the polymer PA66 (density ~1.14 g/cm$^3$) and the diameters of the carbon fibres (diameter=7-8 µm) are rather low, the individual fibres can be clearly seen. The right picture of Figure 2 shows a three-dimensional visualisation of the XCT-data of a carbon-fibre reinforced polymer (CFRP) from the aeronautical industry. Carbon fibres, resin and pores can be distinguished. The segmented carbon fibres are shown in green, the resin in red and the pores in blue.

The left picture in Figure 3 shows an XCT cross-sectional picture of a basaltic fibre-reinforced polymer measured with a voxel size of (10 µm)$^3$ by the RayScan 250E device. The basaltic fibre bundles can be clearly seen and analysed. The right picture of Figure 3 shows an XCT cross-sectional picture of talcum-filled polypropylene. Due to the high resolution and high coherence phase contrast, effects occur at the edges of the particles and polymer [4] resulting in an upward and downward overshooting of the grey values. The individual talcum particles can be clearly seen in Figure 3.

Fig. 3. Cross-sectional XCT picture of a basaltic fibre-reinforced polymer measured with the Ray Scan-device with a voxel size of (10 µm)$^3$ and a cross-sectional XCT picture of PP-talcum measured with the Nanotom device with a voxel size of (0.5 µm)$^3$.

Figure 4 shows cross-sectional XCT pictures (axial, frontal and lateral) of polypropylene filled with mica (glimmer) particles (left pictures) and 3D-visualisation of the mica particles (right picture) measured with sub-µm XCT. The cross-sectional pictures show the distribution of the mica particles and some pores, which have dark grey values. Some of the mica particles agglomerate resulting in big particles. The right picture in Figure 4 shows the segmented big mica particles in three dimensions. The position and distribution of the agglomerations can thus be analysed.
The left picture of Figure 5 shows a cross-sectional XCT picture of polypropylene fibres with 4% cellulose particles measured with a sub-µm-XCT (voxel size (0.7 µm)³). The cellulose particles have a higher density (1.5 g/cm³) than the polypropylene (0.9 g/cm³) and appear therefore as bright particles within the fibres. The right picture of Figure 5 shows a cross-sectional XCT picture of a polyurethane foam filled with cellulose particles. This material is a new moisture-absorbing wood-plastic composite used for special mattresses. The cellulose particles within the polymeric foam structure are clearly visible and can be differentiated from the polymeric matrix. It can also be seen that the grey value at the polymer-air interface is much higher than within the polymer. This can be explained by a rather strong phase contrast effect caused by the small focal spot size and the high resolution of the XCT-measurement [4,7].

Figure 6 shows a cross-sectional XCT picture of polypropylene with cellulose fibres. The cellulose fibre can be clearly differentiated from the polymeric matrix. In contrast to glass and carbon fibres the cellulose fibres are quite bent and have rather irregular shapes. The orange arrow marks a crack within the polymeric sample.
Figure 7 shows a comparison between a glass-fibre and a cellulose-fibre reinforced polymeric samples. Cross-sectional XCT pictures of polypropylene with glass fibres and with cellulose fibres are presented. Both samples were produced by injection moulding. The picture demonstrates that the glass fibres are not present at the surface regions of the bottom of the part, whereas the cellulose particles are equally distributed and are also present in these regions at the bottom of the part.

Figure 8 shows cross-sectional XCT pictures of wood-plastic composites. The cross-sections of the oriented strand board (OSB) show individual wood particles and pores. The wood particles have sizes up to a length of 20 mm. The line bottom right shows the density distribution of the oriented strand board along the line shown in Figure 8. In the right picture of Figure 8 a wood-plastic composite with a lower wood content and smaller wood particles is presented. The size and distribution of the individual wood particles and the pores are clearly visible and can be analysed.
3.2 Extraction of quantitative values from the XCT-data

From the XCT-data quantitative data can be extracted. For instance, the XCT-data of glass-fibre reinforced polymer can be processed to extract fibre orientation and fibre length distribution function [6]. The XCT-data is processed by several consecutive steps: pre-processing, segmentation, medial axis extraction, individual fibre extraction including cluster analysis and final fibre characterisation [6]. Figure 9 shows fibre length distribution functions of injection moulded polypropylene reinforced with 10 %, 30 %, 40 %, 50 % and 60 %. It is clearly recognisable that the fibre length distribution function is shifted to lower values with increasing fibre content. The higher the fibre content the more fibres will be broken during the die injection moulding process. Further details can be found in [6] and [8].
Not only can fibre lengths be measured but also particle sizes can be extracted. This is shown in Figure 10, where the particle diameter distribution function for cellulose particles of the polyurethane foam from Figure 5 is presented. Due to the limited CT-resolution particles with a diameter below 10 µm were not considered. It can be seen that the maximum distribution function is at small diameters and the particle diameters reach values up to around 80 µm. The shape of the distribution function resembles an exponential function.

![Particle Diameter Distribution](image)

**Fig. 10.** Cellulose particle diameter distribution for the PU-foam with cellulose particles.

### 4. Conclusions

We have presented high resolution XCT results, measured with cone beam desktop devices, for polymers reinforced with different fillers measured with voxel sizes down to (0.5 µm)$^3$ and discussed what is possible. The fillers used were fibres and particles made of inorganic, cellulose-based and polymeric materials. From the XCT-data quantitative XCT-data like fibre orientation, filler content, three-dimensional distribution of the fillers and filler size distribution function can be extracted. By using these results, polymer processing methods like injection moulding and the physical properties of the final materials can be analysed.

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### References


