Properties of sustainable composites based on bio-based polyamides and recycled carbon fibers under static and cyclic loads

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Properties of sustainable composites based on bio-based polyamides and recycled carbon fibers under static and cyclic loads

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Abstract. The demand for high-performance materials increases in the last decade and sustainability of materials becomes more important as well. Different bio-based polyamides of type x.10 were compounded with 30wt% recycled carbon fibers to achieve a material with high mechanical properties with low CO₂ emissions. The micro structure was evaluated by using high-resolution tomography (voxel size under 1µm) and scanning electron microscopy (SEM). Mechanical tests were done to characterize the quasi-static and fatigue performance of the composites. The carbon fiber composites show a significant increase of Youngs’ Modulus and tensile strengths compared to the neat polyamides. The fatigue strength measured by an increasing stress fatigue test is approx. 67% of the tensile strength of each studied composite. Besides the short carbon fibers (mean value about 80µm) larger impurities from the recycled carbon fibers were observed. Due to that and the fiber recycling process, the tensile strengths of these sustainable composites are lower compared to composites with virgin carbon fibers and approximately on the level of glass fiber reinforced polyamides.

1. Introduction
The technical requirements of fiber reinforced composites increased in the last few decades to compete with steels in structural applications e.g. in the aircraft industry. Thermosets resins are widely used in this field as matrix material. The amount of carbon fiber reinforced thermosets is about 50% by weight in a modern aircraft [1]. Due to this the amount of high value production waste and components with drawbacks in reuse increases, too. In addition to that, sustainability becomes more and more important in all areas of plastic industry. Recycling is an opportunity and an important issue in all areas of industries. To enhance the sustainability of parts based on carbon fiber reinforced thermosets a recycling method based on pyrolysis is commercially available and provides short carbon fibers [2][3][4]. They can be used as a reinforcement for thermoplastic materials in injection molding or extrusion processing. The recycled carbon fibers (rCF) offer higher mechanical properties compared to glass fibers and a lower price and carbon foot print compared to virgin carbon fibers [2].

Different papers about rCF and composites with rCF are already available. E.g. Feng et al [5] found that the tensile strength and the Youngs’ modulus of composites based on PA 6 with rCF can be increased by a fiber treatment with DGEBA epoxy resin. A maximum tensile strength of 170 MPa and a Youngs’ modulus of 20 GPa were achieved in this investigation.

Composites based on polyamides and short carbon fibers with a fiber content of 30% by weight are commercially available and structural parts like gearings and lever-arms are made of those composites [6]. Therefore, for such applications cyclic loads become more important and has to be determined.
There are just a few studies focusing on bio-based polymers using short carbon fibers, but in terms of sustainability bio-based plastics are also in the focus of interest. Up to now, the focus of bio-based plastics is mostly on packaging and single-use products. There are just a few applications using engineering bio-plastics such as bio-based polyamides. The author [7][8] studied different polyamides with glass and cellulose fibers and found approx. 7.2 GPa Youngs’ modulus for PA 6.10 and PA 10.10 with 30wt% E-glass fibers and a tensile strength of about 130 MPa for PA 10.10 30GF and about 140 MPa for PA 6.10 30GF. Besides this, composites with regenerated cellulose fibers show a higher temperature sensitivity and therefore higher requirements regarding processing are necessary [8][9]. Compared to glass fibers higher impact properties can be achieved with the regenerated cellulose fibers as a reinforcement in several matrix materials and also bio-based polyamides [7]. Benefits in reduced water uptake and carbon footprint are also known for the bio-based polyamides [10].

The aim of this study is to compound bio-based polyamides with rCF and investigate the mechanical properties of the sustainable composites. The characterization of the microstructure should give an impression about the effect on the mechanical properties.

2. Materials and Methods

2.1. Materials

The bio-based polyamides 10.10 and 6.10 from the company Evonik Industry and the bio-based polyamide 4.10 from the company DSM were used as a matrix material. All bio-based polyamides are based on castor oil. Polyamide 10.10 is fully bio-based, the polyamide 6.10 approx. 62% and the polyamide 4.10 71% bio-based. From polyamide 10.10 to 6.10 to 4.10 the tensile strength, tensile modulus and heat distortion temperature increases. All investigated matrix materials show significant lower carbon footprints compared to common petrol-based polyamides with similar specifications [10].

Recycled short carbon fibers (chopped) with a treatment for polyamides from the company CarboNXT were used as fiber reinforcement. A stabilizer package for polyamides (Polyad ®201) from the company PolyAd Services was used to protect the composite against processing and service degradation.

2.2. Composites Preparation

The materials were compounded with a co-rotating twin-screw extruder from the company Leistritz with a screw diameter of 18 mm and a L/D ratio of 40. The additive was fed together with the polymer in the main barrel and the fibers were fed directly in the melt by using a side feeder. The screw configuration was as described in [8] and the processing temperatures were set to 20°C above the melting point of each matrix material. The strand was pelletized into 3 mm granules.

After compounding the dried granules (less than 0.2% moisture) were processed with an injection moulding machine C320 Golden Edition from the company Arburg with a screw diameter of 25 mm. The mold temperature was set to 80°C and the melt temperature 20°C above the melting point.

2.3. Characterization

All composites were characterized at dry state.

The tensile test was performed according to ISO 527 and a fatigue test with increasing stress was performed to characterize the mechanical properties. The fatigue test starts with 30 MPa (upper stress level, significantly under the tensile strength or yield strength) and the stress increases every 2.5k cycles by 2.5MPa. The frequency was set to 5 Hz and the stress ratio (lower stress to upper stress) was 0.1. The and cycles to failure and the failure stress were evaluated.

Additionally, scanning electron microscopy (SEM) and high-resolution X-ray computer tomography (voxel size 0.9 µm) were performed to characterize the microstructure of the composites. The SEM images were done on the break surface after tensile test of polyamide 10.10 specimen with 30wt% rCF. The X-ray tomography was performed on a cutout at the center in longitudinal direction
of a polyamide 10.10 test specimen. The half the volume of the cross-section was measured in respect to a homogeneous material behavior and to achieve a more symmetric material thickness during the measurement (in direction of rotation). The measured volume was 10 mm by 4 mm by 5 mm.

3. Results

3.1. Tensile Properties

The results of the tensile test are shown in Figure 1. As expected, the strength and Young’s modulus of neat PA 4.10 is higher compared to PA 6.10 and PA 10.10 due to their chemical structure and number of amine-groups in the macromolecule. The composites follow the trend of the matrix materials and the rCF lead to a significant increase in strength and Young’s modulus. A factor of about 20 was observed for the Young’s modulus compared to the neat matrix material (up to 25 GPa for PA 4.10 with 30wt% rCF). This is a factor of 3 compared to composites with 30wt% glass fibers (see [7]). The tensile strength is slightly higher compared to glass fiber reinforced composites (see [7]) and approx. 30% lower compared to composites with virgin carbon fibers. The tensile strength of dry-state PA 4.10 30GF is 170 MPa, the Youngs’ modulus is 9.5 GPa according to the suppliers’ datasheet. A commercially PA 4.10 with 30wt% carbon fibers is not available, but comparable to the bio-based PA 4.10 is the common petro-based PA 6.6. In datasheets for PA 6.6 with 30wt% virgin carbon fibers a tensile strength up to 300 MPa and a Youngs’ modulus up to 24.5 can be found.

![Figure 1: Youngs’ Modulus (left) and tensile strength (right) of all composites and the neat matrix materials](image)

3.2. Fatigue Properties

Figure 2 show the failure stress and the cycles to failure of all composites and the unreinforced bio-based polyamides. It can be studied that the failure stress of all composites ranges between 66% and 68% of the quasi-static tensile strength. This does not correlate to the failure stress of the unreinforced polyamides. For the polyamides the highest relative stress was observed for PA 4.10 compared to PA 6.10 and 10.10. This might be caused by the higher heat distortion temperature of PA 4.10 and therefore the resistance against the warming due to the cycle loads. Parallel to the higher failure stresses the cycles to failure increases as well. For the PA 4.10 more than 100k cycles occur before failure. The lowest cycle number was observed for the PA 10.10 composites (71k cycles). Unreinforced polyamides achieve the lowest number of cycles to failure starting from 9k cycles for PA 10.10 to 11k cycles for PA 6.10 and 28.6k cycles for PA 4.10.
3.3. Scanning Electron Microscopy (SEM)

The microstructure of a representative break surface of a PA 10.10 30rCF composite is shown on Figure 3. The fiber distribution is homogeneous and the fiber matrix adhesion is high based on visual inspection. Besides the single filaments also larger agglomerates and larger impurities can be seen on the break surface. The larger impurities cannot be allocated with fiber-like material. The fiber agglomerates could be a result of the fiber treatment for polyamides after pyrolysis. Obviously, those were not fragmented during compounding.

3.4. High Resolution X-ray Computer Tomography

Figure 4 shows the microstructure of a cutout of a PA 10.10 specimen with 30wt% rCF including fibers and different impurities. The impurities are significant larger compared to the fibers. The mean value of the fiber length is approx. 80 µm and the diameter is about 8 µm. The impurities have a length of approx. 200 µm and also a significant higher diameter without the typical carbon fiber shape. Unreinforced polyamides do not show these impurities. Based on the images, it can be concluded, that the impurities are mostly no fiber agglomerates and must be contained in the rCF.
4. Conclusions

Fully and partly bio-based polyamides were compounded with recycled carbon fibers and characterized. The composites allow to achieve outstanding properties in short fiber reinforced polyamides for injection molding applications with respect to sustainability. The significant higher Youngs’ modulus and the lower density of the composites compared to glass fiber reinforces polyamides lead to benefit for light weight parts. The lower tensile strength of the composites with rCF compared to composites with virgin carbon fibers are based on the recycling process. The large impurities act as failures in the microstructure of the material. The fatigue performance of the composites measured by a fatigue test with increasing stress show a failure stress of about 67% of the tensile strength of each composite. Caused by the higher properties of PA 4.10 the mechanical properties of PA 4.10 composites are higher compared to PA 6.10 and PA 10.10 composites.

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