Melt-spun polyamide-filaments with TiO_2-nanoparticles: influence on thermal conductivity and tensile strength

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One solution for increasing the thermal conductivity of thermoplastics is adding nanoparticles to the polymer. Various titanium dioxide particle types are tested, which differ in their surface area. Also the amount of TiO_2 is varied between 0 and 20 wt-%, because a compromise is needed between increased thermal conductivity and reduced strength and rigidity caused by increasing the concentration of particles. Furthermore, the way of compounding and the influence of dispersant on the particle dispersion in the polymer matrix are examined. Inter alia it could be determined that an amount of 20 wt-% TiO_2 in polyamide 6 leads to an increase of thermal conductivity of 26 %. It can be shown that polyamide 6-filaments with 5 wt-% TiO_2 can be melt spun. The mechanical properties of these modified filaments are still high enough for further textile treatment.
Key words: *Titanium dioxide, polyamide 6, thermal conductivity, filaments, melt spinning process*

1. Introduction

Light-weight construction becomes nowadays more and more important. Especially in the automotive industry the prevailing motto is: The lowest possible weight at constant or better mechanical properties. Reducing weight often means reducing costs. In order to put this in effect, Fibre reinforced thermoplastic composites (FRTCs), so called organic sheets, are developed. Therefore, thermoplastic fibers are intermingled with reinforcement fibers and processed into textile fabrics. Subsequently, consolidation and forming follow in a hot press under the influence of temperature. In Fig.1 the complete process chain is shown. The approach, which is already reached at ITA, is pointed out.

For an optimal consolidation, the temperature distribution needs to be homogenous throughout the organic sheets [1]. Too high adjusted temperature leads to damage in the polymer structure. A lower temperature requires more dwell time in the hot press. This step is actually the bottleneck of the process chain. The qualitative temperature distribution throughout the parts thickness is presented in Fig.2.

With a faster process comes a reduction of costs. While the weight reduction enforces higher energy efficiency thus decreasing operating costs, the main driver for research in this field are governmental regulations. For example the European Union aims to reduce the production of CO_2 by 20 % until 2020 in comparison to 2009. In order to reach this goal it is important to create a sustainable and economic way of mobility and transportation [2]. These organic sheets have the potential to link low weight with a large scale, series production. One solution for shorter cycle times is investigated at ITA. Nanoparticles are added to the polymer to increase its thermal conductivity.

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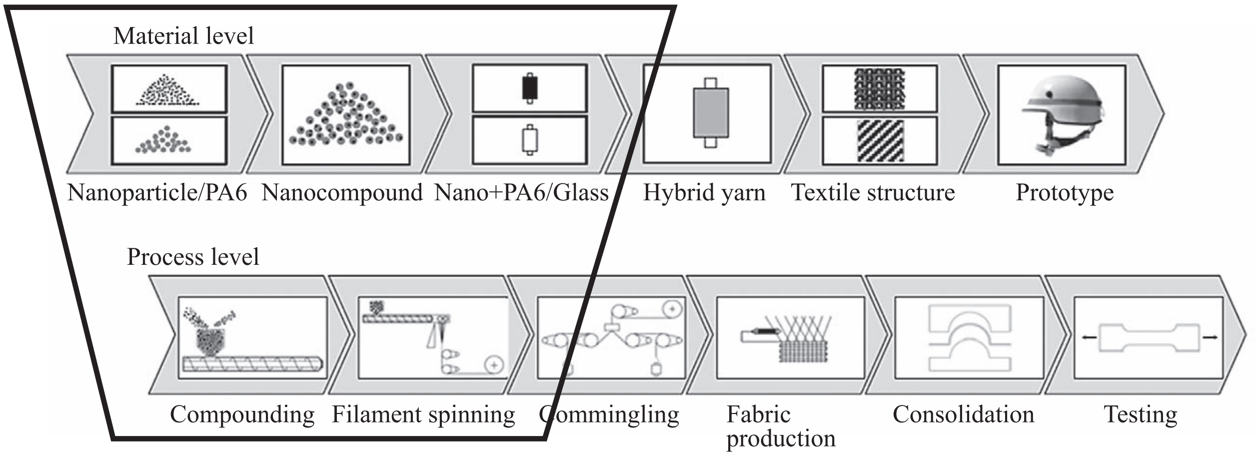

Fig.1 Approach and process chain of producing organic sheets at ITA

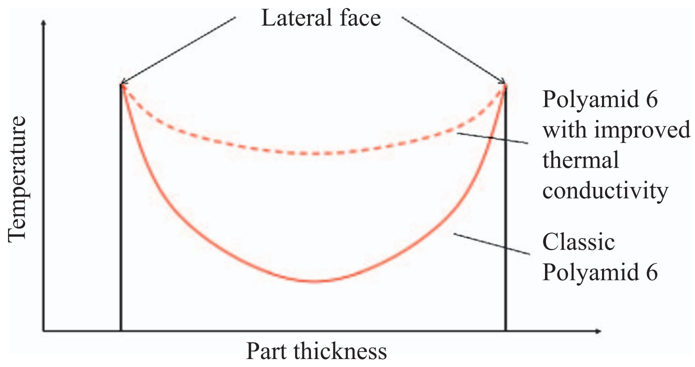

Fig.2 Qualitative temperature gradient

2. Materials and methods

This chapter focuses on the experimental part of the mentioned research and presents the materials and applied methods. The thermal conductivity of the used, pure materials and the surface area of the TiO_2 nanoparticles are shown in Tab.1.

Polyamide 6 (PA 6), with a relative viscosity of 2.4 is used as matrix polymer. It was compounded as powder and granulate in order to study compounding effects. As fillers TiO_2 nanoparticle types RM 220 and RM 300 are used, which only differ in particle size and therefore in surface area. Furthermore a dispersion aid, Brüggolen P 130, is used in some trials to investigate if there is a significant positive effect on increasing the thermal conductivity or on the spinnability in the melt spin process.

The materials are compounded with a twin-screw-extruder Lab-Compounder KEDSE 20 / 40 D from Brabender GmbH & Co. KG, Duisburg, Germany at an extrusion temperature of 260 °C. The amount of fillers was varied throughout the testing and can be withdrawn from Tab.2.

The thermal properties of the compounds were investigated by capillary rheology (Fig.3).

This testing method induces a defined heat flow into the material and measures the time needed to increase its heat by a predefined amount. With the gathered data points the thermal conductivity can be calculated by the following equation (1):

$$\lambda = \frac{Q \cdot k}{4 \cdot \pi \cdot l} \cdot \frac{\ln \frac{t_2}{t_1}}{T_2 - T_1} \qquad (1)$$

where λ is the thermal conductivity, Q resembles the amount of energy fed into the sample, k is a constant of the capillary rheometer, l the length of the heat wire, t_1 and t_2 are the first and

Tab.1 Properties of the used materials

Material	Thermal conductivity - λ [W/mK]	Surface area - S [m²/g]
Polyamide 6 (PA 6)	0.245	-
TiO_2 RM 220	12.0	60
TiO_2 RM 300	12.0	70

Tab.2 Filler amount of the different compounds

Polymer	Filler	Filler amount [wt.%]
PA 6	TiO_2 RM 220	4
PA 6	TiO_2 RM 300	4
PA 6 - powder	TiO_2 RM 300	4
PA 6	TiO_2 RM 220	10
PA 6	TiO_2 RM 300	10
PA 6	TiO_2 RM 300 + P 130	10 + 0.1
PA 6 - powder	TiO_2 RM 300	10
PA 6	TiO_2 RM 300	14
PA 6	TiO_2 RM 220	14
PA 6 - powder	TiO_2 RM 300	14
PA 6	TiO_2 RM 300	20
PA 6	TiO_2 RM 300 + P 130	20 + 0.2
PA 6	TiO_2 RM 220	20
PA 6 - powder	TiO_2 RM 300	20

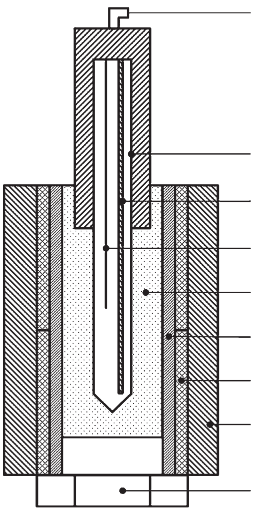

Fig.3 Schema of capillary rheology

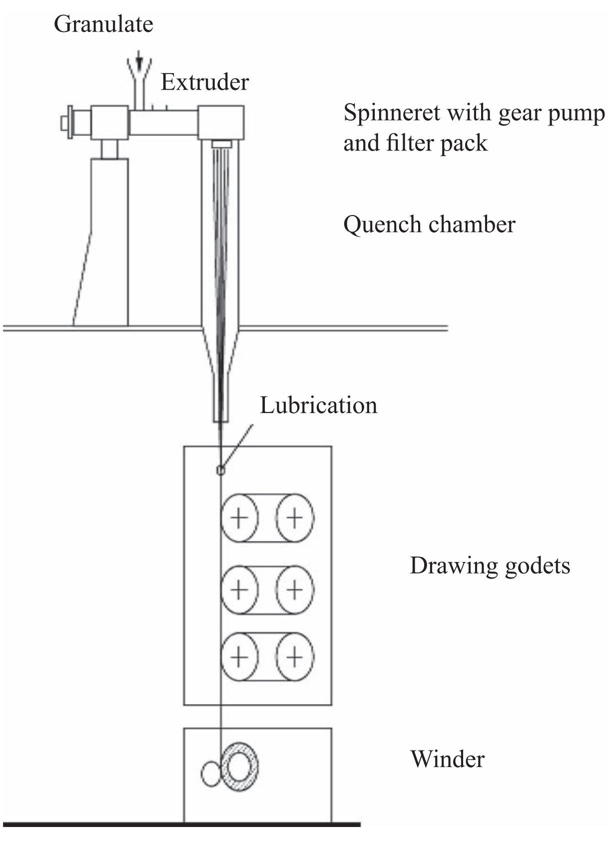

Fig.4 Shema of melt spinning

second point of time measurement, T_1 and T_2 the first and second point of temperature measurement [3].
The modified filaments, with an amount of 5 wt-% TiO_2, are spun on a melt spinning plant at ITA, which can be seen in Fig.4. A 96-hole-spinneret with diameters of 25 µm is used. The yarn is produced as LOY to keep the shrinkage, which may occur afterwards, as low as possible. The mechanical properties are investigated by uniaxial tensile test to control if the mechanical properties, despite the presence of particles, are suitable for further textile processing.

3. Results

This section describes the results of the beforehand mentioned investigations. Firstly the effect of compounding between using TiO_2 powder and PA 6 powder or granulate is described.
This is followed by an analysis of the modification of different TiO_2 types and an additional use of dispersion aid. Regarding the results of the granulate based compounds a constant rise of the thermal conductivity with the TiO_2 content can be detected (figure 5). The rise of the powder based compounds is not significant. It can be seen that the form of the polymer does not have a significant positive influence on the thermal conductivity. Furthermore the small deviation of the measurements of the granulate pattern implies a reproducible correlation.
The following graph shows the results of the measurement of the thermal conductivity for the different TiO_2 types RM 220 and RM 300 (Fig.6). Overall a rise of the thermal conductivity with an increasing amount of TiO_2 can be observed. This rise seems to be independent from the type of TiO_2 used.
Hence, the melt spinning trials are pursued with only one type of TiO_2, the RM 220. The experiments also show that the use of dispersion aid has no significant influence on the thermal conductivity. Anyhow, P 130-containing nanomodified material is used is some spinning trials expecting a positive effect on the spinnability. The trials show that it is possible to melt spin filaments containing 5 wt-% TiO_2 with a process, which is stable for 12 hours at least. The mechanical properties are shown in tab.3. The tensile strength is lower than in filaments out of pure PA 6, but is still high enough for further textile processing like weaving, for example. The dispersion aid has no significant effect on spinnability or filter life. Admittedly, using a dispersion aid is more expensive and means an additional processing step. Therefore it is not used in following tests.

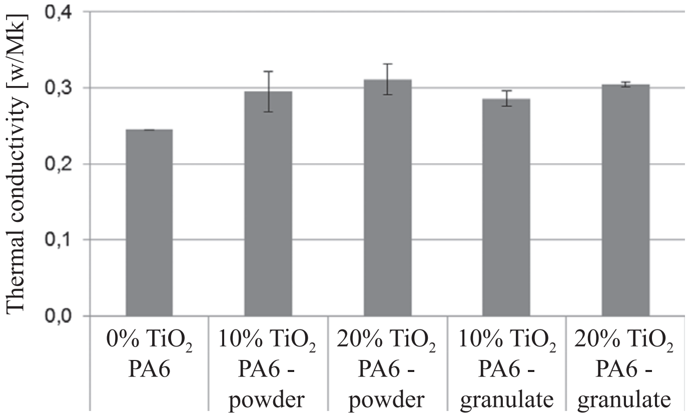

Fig.5 Thermal conductivity of compounds made of TiO_2 and Polyamid 6 as powder and granulate

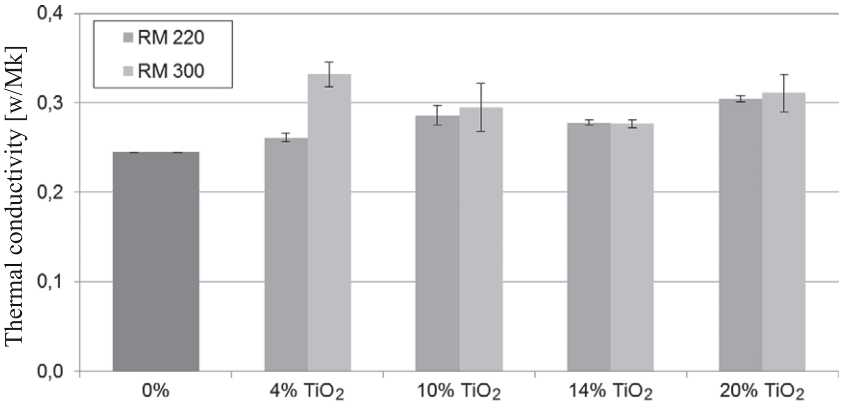

Fig.6 Thermal conductivity of Polyamid 6 compounds containing TiO_2 with different modifications

Tab.3 Mechanical properties of nanomodified und unmodified melt spun filaments

Property	Value	Value
	nanomodified PA6-filaments	unmodified PA6-filaments*
titer	513.4 dtex	484.41 dtex
tensile strength	363.7 cN	812.15 cN
remaining expansion	255 %	244 %

* There is no reference yarn made out of the same PA6 with the same process parameters. The values for pure PA6 shown in Tab.3 are from a similar produced yarn

4. Conclusion

In the following, the results are interpreted towards their influence on thermal conductivity and mechanical properties. Using a powdered thermoplastic instead of a thermoplastic in granulate form has no effect on the thermal conductivity of the compound. Utilizing powdered polymers is more expensive as well as technologically laborious and is therefore not advised. The examinations of different particle types of TiO_2, which are investigated in the next step, show no significant influence on the thermal conductivity. This leads to the conclusion that the surface area does not have a significant effect on the thermal conductivity, as this was the only difference between RM 220 and RM 300. However the high deviation observed for compounds containing 4 % TiO_2 leads to the assumption that the surface area influences the particle distribution and thermal conductivity at low filler amounts. Further investigations are necessary. The experiments have also shown that the use of the dispersion aid Brüggolen P 130 has neither a significant influence on the thermal conductivity nor on the spinnability. Therefore the use is not recommended. Modifying PA 6 with nanoscale TiO_2 leads to an increase of λ. With a filler amount of 20 wt-% the thermal conductivity can be improved by 26 %. The trials have also shown that melt spinning of filaments with 5 wt-% TiO_2 is possible. The process is so stable that further experiments with a higher amount of filler are planned. The mechanical properties are positive as well, so the next steps in the previously mentioned process chain (figure 1) can be approached.

Acknowledgements

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