# Influence of feeding conditions in twin-screw extrusion of PP/MWCNT composites on electrical and mechanical properties

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# Abstract

The influence of feeding conditions of multiwalled carbon nanotube (MWCNT) materials, namely Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000, into polypropylene (PP) was investigated with respect to achieve suitable nanotube dispersion, high electrical conductivity, and good mechanical properties. Both MWCNT materials were fed at selected concentrations either in the hopper of the twin-screw extruder or using a side feeder under otherwise identical extrusion conditions (rotation speed, throughput, temperature profile) using a Berstorff ZE25 twin-screw extruder. Afterwards, injection molding was performed under identical conditions. The results indicate that the more compact Baytubes<sup>®</sup> C150P agglomerates should be added into the hopper as dispersion assessed by light microscopy is better, electrical resistivity measured on compression and injection molded samples is lower, and elastic modulus, yield strength and impact strength are higher and as compared to side feeding. On the other hand, for the more loosely packed Nanocyl<sup>TM</sup> NC7000 agglomerates addition using the side feeder leads to better dispersion, lower electrical resistivity, and higher mechanical properties.

*Keywords:* A. Carbon nanotubes, Nano composites, Polymer-matrix composites (PMCs); B. Electrical properties; D. Optical microscopy, Dispersion

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## **1. Introduction**

Advanced materials containing carbon nanotubes (CNTs) are being developed for many industries and research on CNT-based polymer composite applications is currently of great interest. The research focus is to transfer the mechanical [1, 2], thermal [3, 4] and the unique electrical properties [5, 6] of CNTs into the polymeric matrix to substitute for traditionally used fillers such as carbon black, or to generate new or improved property combinations. Polymer based CNT composites can be used for applications in the fields of electromagnetic interference shielding [7, 8], electrostatic dissipation [9] and electrically conductive materials. In addition, enhanced mechanical properties and thermal stability [10] can be achieved as well as improved thermal conductivity. The full potential of CNT composites in industrial applications is not yet fully realized due primarily to the fact that generating good CNT dispersion and distribution in the polymer matrix is quite difficult [11]. The majority of previous studies using melt mixing to disperse CNTs into polymers were done using small-scale mixing and have illustrated that melt processing conditions must be optimized to achieve good CNT dispersion in the composites [12-17]. Andrews et al. [16] have shown for nanofibers in different matrices that the highest energy input in a batch mixer, achieved either by high rotation speed or long mixing time, led to the best dispersion but also decreased nanofiber length most severely. Kasaliwal et al. [12] indicated that mixing at high melt temperature and high mixing speed can yield composites with high degree of dispersion observed by light microscopy at relatively low mixing energy input with a minimum damage for the CNTs.

Literature in the field of laboratory-scale melt mixing of CNTs into thermoplastic polymers using extruders is sparse [18-25]. Most studies are made under constant mixing conditions and do not consider the influence of melt mixing conditions on CNT dispersion or electrical percolation. Villmow et al. [18] investigated the influence of extrusion conditions on the dispersion of multiwalled carbon nanotubes (MWCNTs) in a polylactide (PLA) matrix using twin-screw extrusion whereas the nanotubes were added together with the polymer in the hopper of a Berstorff ZE 25 extruder. Processing conditions e.g. screw configuration, screw speed, and temperature profile, influence CNT dispersion and distribution during both the masterbatch production step and during dilution. A rotation speed of 500 min<sup>-1</sup> that still ensures a certain residence time of the melt combined with a screw profile containing mainly mixing elements were found to be best in dispersing and distributing MWCNTs in the PLA matrix during both masterbatch production as well as the dilution step. Villmow et al. [20] discussed the influence of screw configuration, rotation speed, and throughput on the residence time and specific mechanical energy (SME) and the resulting macroscopic CNT dispersion in polycaprolactone (PCL)-based masterbatches. As in Reference [18], nanotubes and polymer pellets were fed together into the hopper of a Berstorff ZE 25 extruder. The best dispersion was found for an extended screw with L/D=48, a high rotation speed of 500 min<sup>-1</sup> and a low throughput of 5 kg/h. In a recent study by McGlory et al. [6] the influence of screw speed during twin-screw extrusion on the electrical percolation threshold of polystyrene-MWCNT was investigated. The dispersion was best at 100 rpm, whereas also lower dispersion qualities achieved at 20 and 70 rpm lead to comparable electrical percolation thresholds between 1 and 3 wt% addition. The highest mixing speed of 150 rpm led to worse dispersion and significantly higher percolation threshold.

The influence of feeding conditions in melt mixing using twin-screw extrusion on dispersion and mechanical properties has been investigated for composites based on other nanoparticles e.g. organoclay or aluminum oxide [26, 27], however for CNT based composites no reports could be found.

Carbon nanotubes from diverse producers can differ in their characteristics such as length, diameter, purity, curvature, agglomerate density and strength, and surface functionalization. Krause et al. [28] reported the relationship between the energy input needed to disperse different multiwalled carbon nanotube (MWCNT) agglomerates in aqueous surfactant solutions and the bulk density and agglomerate deformability of these MWCNT materials. According to these investigations, CNT materials with higher bulk density need more energy input to establish stable aqueous dispersions. In addition, a correlation was found between the dispersability in aqueous dispersions and the dispersion after melt mixing in a PA12 matrix [29] and in a PC matrix [30].

Based on these results, optimization of extrusion melt mixing conditions for different nanotubes might be necessary in order to achieve optimal dispersion in a particular polymer. In other words, processing conditions such as screw configuration, mixing speed, temperature profile, throughput, and the way of filler feeding have to be adapted for the respective type of MWCNT. In the investigations described herein, a variation of feeding conditions for two types of commercially available MWCNT materials with different bulk densities, namely Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000, was performed. MWCNTs were incorporated in impact-modified polypropylene (PP) containing talc commercially used in automotive applications. Composites having different MWCNT content were prepared with the aim to achieve high electrical

conductivity at low nanotube loadings. All extrusion runs were realised under otherwise identical melt mixing conditions using a twin-screw Berstoff ZE-25 extruder so that the effects of the MWCNT feeding location, either in the hopper or using a side feeder, could be studied selectively. Optical light microscopy, electrical conductivity and selected mechanical measurements were performed.

## **3** Experimental

#### **3.1 Materials**

Impact modified polypropylene (PP) filled with 30 wt% talc having a melt temperature range of 160 – 165°C and a melt flow index of 13 g/10 min (ISO 1133) was used as the matrix. Two different commercially available multiwalled carbon nanotube materials were employed, namely Baytubes<sup>®</sup> C150P (Bayer MaterialScience AG, Leverkusen, Germany) and Nanocyl<sup>TM</sup> NC7000 (Nanocyl S.A., Sambreville, Belgium). According to investigations presented in [31] the Baytubes<sup>®</sup> C150P nanotubes have mean diameters of 10.5 nm and a mean nanotube length of 770 nm. The Nanocyl<sup>TM</sup> NC7000 nanotubes were measured to have mean diameter and length of 10.0 and 1341 nm, respectively. According to the suppliers, the bulk density of Baytubes<sup>®</sup> C150P is 120-170 kg/m<sup>3</sup> [32] whereas that of Nanocyl<sup>TM</sup> NC7000 is reported to be 60 kg/m<sup>3</sup> [33]. Measurements of the stress needed to compress as received single agglomerates to 25% deformation resulted in values of 0.64 MPa for Baytubes<sup>®</sup> C150P and 0.39 MPa for Nanocyl<sup>TM</sup> NC7000 [28] illustrating an easier deformability of the latter MWCNT agglomerates.

#### **3.2 Processing**

To examine the electrical percolation thresholds, composites were produced using a DSM Xplore twin-screw microcompounder (DSM Xplore, Netherland) with a chamber volume of 15 cm<sup>3</sup>. PP granules and MWCNT powder were premixed and fed together in the hopper. Mixing conditions were 190°C melt temperature, 150 rpm mixing speed, and 5 min mixing time. Extruded strands were compression molded (200°C, 50 kN) into plates (60 mm diameter, 0.3 mm thickness) using a hot press (Model PW 40 EH, Paul Otto Weber GmbH, Germany).

For twin-screw extrusion, an intermeshing co-rotating twin-screw extruder ZE25 (Berstorff, Germany) having a distributive mixing screw configuration with an L/D length of 48 (similar to screw SC-5 in [20]) was used. The MWCNTs were fed gravimetrically either in the hopper with the polymer granules or using a side feeder attached at 14 D. An increasing temperature programme from 180°C to 200°C, a screw speed of 500 min<sup>-1</sup>, and a throughput of 10 kg/h were used. The compounded pellets were compression molded (200°C, 50 kN) into circular plates (60 mm diameter, 0.3 mm thickness) using a hot press (Model PW 40 EH, Paul Otto Weber GmbH). Injection molding using typical PP molding conditions was performed using an injection molding machine Ergotech 100/420-310 (Demag, Germany) using a two-cavity mold, fed by a flash gate, using a melt temperature of 240°C and a mold temperature of 40°C. For electrical tests plates of 80 mm x 80 mm x 2 mm were injection molded (Fig. 1) and dogbones with dimensions of 40 mm x 150 mm x 4 mm and bars of 10 mm x 80 mm x 4 mm were molded for tensile and impact tests, respectively.

## 3.3 Characterization

#### 3.3.1 Electrical resistivity

The electrical resistivity of the compression and injection molded plates was determined according to the standards ASTM D 4496 and ASTM D 257. Surface as well as volume resistances were measured and converted to resistivity values. At least six compression molded samples and two injection molded samples were measured on both sides to obtain the geometric mean values of resistivity which together with the standard deviation as error bars are shown in the plots. Measurements on the pressed plates or injection molded samples with resistances >10<sup>7</sup> Ohm were done using a Keithley 8009 Resistivity Test Fixture (open symbols in the graphs). For resistances lower than  $10^7$  Ohm for the pressed plates, strips (5 mm x 55 mm x 0.3 mm) were cut from the plates and measured using a 4-point test fixture (external source electrodes spacing 16 mm and measuring electrodes spacing 10 mm, filled symbols in the graphs). Both devices were combined with a Keithley electrometer 6517A (Keithley Instruments Inc., Cleveland, USA).

The measurements of the injection molded samples with resistances  $< 10^7$  Ohm were performed using a Loresta-GP electrometer in combination with an 4-point ESP-probe (Mitsubishi Chemical Corporation, Japan, external source electrodes spacing 15 mm and measuring electrodes spacing 5 mm). The 4-point probe was located in the middle of the sample either in x-or y-direction (see Fig. 1). Four measurements were performed in each direction and mean values are shown. Thus, volume resistivity is measured through the sample thickness for high resistances (z-direction in Fig.1, through-plane) and mainly in-plane for low resistances.

#### 3.3.2 MWCNT macrodispersion

To study the state of MWCNT macrodispersion within the extruded composites, light microscopy (LM) according to the standard ISO 18553 using an Olympus BH2

microscope in transmission mode was applied. The microscope was combined with a camera DP71 (Olympus Deutschland GmbH, Hamburg, Germany) and thin sections of 10  $\mu$ m thicknesses were investigated. Thin sections were prepared from extruded pellets using a Leica RM 2155 microtome (Leica Microsystems GmbH, Wetzlar, Germany). The agglomerate area fraction was determined from the LM images using the image analysis software ImageJ Version 1.430 by calculating the ratio (in %) of the area of agglomerates A (according to the ISO 18553 standard only agglomerates with circle equivalent diameters > 5  $\mu$ m are regarded) to the total area of the image A<sub>0</sub>. For quantification at least 15 cuts were investigated for each sample and the standard deviation between the 15 cuts is mentioned.

#### 3.3.3 Investigation of MWCNT's pristine morphology

Scanning electron microscopy (SEM) was performed on the pristine MWCNT materials using an Ultra Plus Field Emission Gun Scanning Electron Microscope (FEG-SEM, Carl-Zeiss AG, Oberkochen, Germany). The MWCNT materials were deposited in their as received dry state on a double-sided adhesive copper tape and examined using a SE2detector.

#### 3.3.3 Measurements of mechanical properties

To compare the mechanical properties of the differently prepared extruded composites, the tensile properties ( $E_t$  Young' modulus,  $\sigma_Y$  yield stress,  $\sigma_M$  maximum stress,  $\sigma_B$  stress at break,  $\epsilon_Y$  strain at yield point,  $\epsilon_M$  strain at maxium stress,  $\epsilon_B$  strain at break) and the notched Charpy impact strength at 23°C ( $a_{cA+23}$ ) were measured according to DIN EN ISO 527-2/1A/ 50 and ISO 179/1eA, respectively.

### **4 Results and Discussion**

#### 4.1 MWCNT morphology

Beside the characteristics of single nanotubes such as length, diameter, purity, waviness and functionalization, the morphology of the nanotube agglomerates is of interest for their dispersion behavior. Figure 2 show significant differences in the SEM images of the two as-received nanotube powders. A combed-yarn structure was observed for Nanocyl<sup>TM</sup> NC7000 whereas for Baytubes<sup>®</sup> C150P a bird nest structure was found. The agglomerate structure of Baytubes<sup>®</sup> C150P seems to be very compact as compared to the more open structure of Nanocyl<sup>TM</sup> NC7000. This finding correlates with the high bulk density of 120-170 kg/m<sup>3</sup> [32] for Baytubes<sup>®</sup> C150P versus the low bulk density of 60 kg/m<sup>3</sup> for Nanocyl<sup>TM</sup> NC7000 [33]. Krause et al. [28] reported that Baytubes<sup>®</sup> C150P agglomerates with a deformation stress (at 25% deformation) of 0.64 MPa needed five times the energy input of Nanocyl<sup>TM</sup> NC7000 (deformation stress of 0.39 MPa) for its dispersion into a stable state in an aqueous surfactant solution. From these results it can be assumed that MWCNT morphology plays a role in achieving good dispersions in melt mixing. In small-scale melt mixing in polyamide 12, Socher et al. [29] showed that Baytubes<sup>®</sup> C150P resulted in worse dispersion and higher electrical percolation threshold than Nanocyl<sup>TM</sup> NC7000. To achieve high electrical conductivity not only the state of dispersion is important, also nanotube length plays an important role. According to Krause et al. [31, 34] the initial length of Nanocyl<sup>TM</sup> NC7000 ( $x_{50} = 1341$  nm) is about twice that of Baytubes<sup>®</sup> C150P ( $x_{50}$  =770 nm). Shortening during processing may also be different with consequences on the nanotubes dispersability and network formation.

### **4.2 Electrical properties**

To investigate the electrical percolation behavior of Baytubes<sup>®</sup> C150P or Nanocyl<sup>TM</sup> NC7000 in PP, composites were prepared in a first set using a DSM Xplore15 twin screw microcompounder. As illustrated in Figure 3, the electrical percolation threshold was found at 0.5 wt% for Nanocyl<sup>TM</sup> NC7000 and between 1 and 2 wt% for Baytubes<sup>®</sup> C150P. This different electrical percolation behavior is likely related to the different dispersabilities [28] and the lower initial length of Baytubes<sup>®</sup> C150P. The comparison between feeding either in the hopper or in the side feeder was performed in the percolation range found in these compression molded samples, namely at 0.5 wt% and 1 wt% for Nanocyl<sup>TM</sup> NC7000 and at 2 wt% and 5 wt% in the case of Baytubes<sup>®</sup> C150P. To compare results at the same nanotube contents, in addition composites with 2 wt% and 5 wt% were prepared for Nanocyl<sup>TM</sup> NC7000.

The influence of feeding position on the electrical volume and surface resistivity of compression molded PP composites containing Baytubes<sup>®</sup> C150P or Nanocyl<sup>TM</sup> NC7000 is shown in Figures 4 and 5. The resistivity values of the composites filled with Nanocyl<sup>TM</sup> NC7000 are slightly lower when the MWCNT were added in the side feeder. For composites filled with Baytubes<sup>®</sup> C150P, resistivity values were lower in the case of adding the MWCNT together with the polymer granules at the hopper. The results were consistent for volume and surface resistivities. These results indicate that the higher shear forces as achieved by feeding the nanotubes together with the unmolten polymer granules in the hopper and the longer residence time in the extruder were favourable for the dispersion of Baytubes<sup>®</sup> C150P in order to create conductive composites. In contrast, for Nanocyl<sup>TM</sup> NC7000 the use of the side feeder into the molten polymer seems to be favourable.

Electrical measurements on injection molded composites (Figures 6 and 7) showed the same tendencies as obtained on the compression molded samples. However, resistivity values were higher as is typical for injection molded vs. compression molded samples [35]. In PP composites with 0.5 wt% nanotubes, no influence of the feeding conditions on resistivity was seen since the skin layer with well-separated and highly-oriented MWCNT is electrically insulating in both cases [19], whereas for the composites with 1 wt% and 5 wt% Nanocyl<sup>TM</sup> NC7000 slightly lower values can be observed when using the side feeder. In the percolation threshold range of the injection molded samples (2 wt% of Nanocyl<sup>TM</sup> NC7000), the use of the side feeder results in a decrease in resistivity by more than two orders of magnitude. In the well percolated range (5 wt%), the difference between both feeding positions is much smaller. The composites with 2 wt% Baytubes<sup>®</sup> C150P were both non-conductive, whereas at 5 wt% only feeding in the hopper resulted in samples with significantly lowered resistivity values indicating beginning percolation.

## 4.2 State of macrodispersion of the CNT fillers

The state of macrodispersion of the remaining MWCNT agglomerates was investigated for all samples. Figure 8 shows the agglomerate area fractions versus the MWCNT content for composites at different feeding conditions whereas Figure 9 shows the comparison of LM micrographs and the related area fractions for the composites with 2 wt% MWCNTs.

When comparing the agglomerate area ratios at constant MWCNT contents (2 and 5 wt%), feeding using the hopper leads to comparable values. However, when using the

side feeder, significant differences in dispersion can be observed (Fig. 8) indicating much better dispersion in case of Nanocyl <sup>TM</sup> NC7000 than for Baytubes<sup>®</sup> C150P.

For Nanocyl<sup>TM</sup> NC7000 lower agglomerate area fractions and smaller agglomerates were found for the samples with CNT feeding into the side feeder. For the 0.5 wt% composite the area fraction decrease from 2.4 % (hopper) to 1.0% (side feeder) and for 1.0 wt% content from 3.8% (hopper) to 2.2% (side feeder). The composites with 2 wt% and 5 wt% Nanocyl<sup>TM</sup> NC7000 show the same dependencies at different feeding conditions. In contrast, the agglomerate area fraction for composites with 2 and 5 wt% Baytubes<sup>®</sup> C150P were lower when feeding was done using the hopper (Figure 8). The observed differences between side feeder and hopper (4.9 % (side feeder) to 2.6 % (hopper) for 2 wt% and 8.6 % (side feeder) to 3.8 % (hopper) for 5 wt%) are higher than those obtained for the Nanocyl<sup>TM</sup> NC7000 composites with 0.5 wt% or 1.0 wt%. Obviously, Baytubes<sup>®</sup> C150P are more sensitive to the feeding position, which again can be related to the much higher bulk density requiring more energy input for agglomerate dispersion.

Summarizing, Baytubes<sup>®</sup> C150P should be added into the hopper together with the polymer granules as this provides higher shear forces and longer residence times. Feeding into the polymer melt obviously does not generate high enough shear forces to disperse the more compact granules by either rupture or erosion like feeding in the hopper and the former results in more and larger remaining agglomerates. Nanocyl<sup>TM</sup> NC7000 material with much lower bulk density seems to require lower energy input and shear forces to disperse it adequately in the polymer melt. Feeding in the hopper

agglomerates by non-molten polymer by the mechanism of the "agglomerate formation" described by Gale [36], and such compacted agglomerates may be more difficult to redisperse in the following extrusion melt mixing process.

#### **4.1 Mechanical properties**

Figures 10 and 11 show typical stress-strain curves and Tables 1 and 2 selected mechanical properties of the injection molded composites with different nanotube loadings. The results illustrate strong dependencies of the mechanical properties on the feeding position for both MWCNTs.

The impact strength of MWCNT composites was reduced after MWCNT addition in comparison to pure extruded PP likely due to non-dispersed primary agglomerates. The decrease increases as more nanotubes were added. In stress-strain diagrams, the talc modified PP shows maximum stress at the yield point at around 5% strain followed by a continuous decrease in stress up to the break of the samples at around 30%. Concerning the Young's modulus, after adding Baytubes<sup>®</sup> C150P generally lower values than for PP were found, whereas the addition of Nanocyl<sup>TM</sup> NC7000 resulted in similar or higher modulus values than for talc filled PP. For Nanocyl<sup>TM</sup> NC7000 incorporation using the side feeder higher values were found than for feeding in the hopper which is in accordance with the better dispersion as observed using light microscopy. A similar trend could be observed for the impact strength showing higher values when feeding Nanocyl<sup>TM</sup> NC7000 in the side feeder. In case of Baytubes<sup>®</sup> C150P, feeding into the hopper resulted in higher values of Young's modulus and impact strength as compared to feeding in the side feeder. The impact strength seemed strongly influenced by the state of nanotube dispersion. For instance the impact strength of PP composites with 2

wt% of Baytubes<sup>®</sup> C150P is about 9.5 kJ/m<sup>2</sup> higher when feeding was done in the hopper (Table 2), whereas the nanotube agglomerate area fraction is by 2.3 % lower than after feeding using the side feeder (Figure 8 and 9). The stress at yield was nearly unaffected by the feeding position when adding Nanocyl<sup>TM</sup> NC7000 (Figure 10) but was 0.8 MPa higher when adding Baytubes<sup>®</sup> C150P using the hopper than adding in the side feeder.(Figure 11). Strain at break decreased clearly with nanotubes addition in case of Baytubes<sup>®</sup> C150P without significant differences between the feeding conditions. For composites with Nanocyl<sup>TM</sup> NC7000 higher strain at break values than for talc filled PP were found after addition of 0.5 wt%, indicating structural or morphological changes in the modified PP induced by the nanotubes. Starting at 2 wt%, a significant decrease in strain at break is observed.

#### **5. Summary and Conclusion**

This study discusses how feeding conditions of two different commercial MWCNT grades, namely Nanocyl<sup>TM</sup> NC7000 and Baytubes<sup>®</sup> C150P, during the melt mixing process in a twin-screw extruder influences the nanotube dispersion of MWCNT in an impact modified polypropylene containing talc. As a consequence of the different states of MWCNT dispersion, the mechanical and electrical properties of the composites were changed.

The influence of the feeding position in twin-screw extrusion of PP was investigated at selected concentrations of Nanocyl<sup>TM</sup> NC7000 and Baytubes<sup>®</sup> C150P materials using a Berstorff ZE 25 co-rotating intermeshing twin-screw extruder with L/D=48. Feeding the nanotubes together with the polymer in the hopper was compared with using a side feeder adapted at L/D=14. Based on the obtained electrical resistivity values of the

composites, measured on compression molded as well as on injection molded samples, in combination with light microscopic investigations of the amount of remaining primary nanotube agglomerates, the following recommendations can be given:

Better dispersion and lower electrical resistivities can be obtained when Baytubes<sup>®</sup> C150P material is added into the hopper together with the polymer granules instead of the side feeder. A higher amount of mixing energy and shear forces can be applied on that material having a relatively high bulk density in order to get suitable dispersion.

For Nanocyl<sup>TM</sup> NC7000 material adding into the side feeder into the already molten polymer is recommended. This nanotube material with relatively low bulk density resulted in better dispersion and electrical properties of the PP composites when fed using the side feeder as compared to feeding in the hopper. When fed in the hopper, the loosely packed nanotube material can be compressed by the nonmolten polymer granules to higher bulk densities and such agglomerates are more difficult to redisperse. These recommendations are in accordance with the advice given by an extruder manufacturer in ref. [37] stating that for CNTs with low bulk density the split feed process and with high bulk density the pre-mixing process is favourable.

The mechanical properties, as Young's modulus and impact strength, were found to be strongly dependent on the state of filler dispersion, being higher in those cases where better nanotube dispersion was obtained.

This study shows that it is necessary to perform individual optimization of extrusion melt mixing for different nanotube materials to achieve good MWCNT macrodispersion. The results clearly indicated that better nanotube dispersion resulted in higher mechanical properties and lower resistivity values of PP based composites. In this study an impact modified PP containing second filler, namely talc, was used in this study which certainly improves the state of dispersion, reduces percolation threshold, and modifies mechanical properties. It can be expected that the differences shown for the two nanotube grades by varying the feeding conditions are also relevant for other types of PP as well as other polymers.

### 6 Acknowledgment

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# **Tables**

MWCNT NC7000	Feeding location	E <sub>t</sub> [MPa]	$\sigma_{\rm Y} = \sigma_{\rm M}$ [MPa]	σ <sub>B</sub> [MPa]	$\varepsilon_{\rm Y} = \varepsilon_{\rm M}$ [%]	ε <sub>B</sub> [%]	a <sub>cA+23</sub> [kJ/m <sup>2</sup> ]
without	-	$1598 \pm 58$	$17.1 \pm 0.1$	$11.4 \pm 0.1$	$4.7 \pm 0.1$	$31.3 \pm 4.2$	$37.6 \pm 1.8$
0.5 wt%	hopper	$1591 \pm 84$	$16.7\pm0.1$	$12.5\pm0.4$	$5.0\pm0.1$	$40.1\pm7.6$	$26.9\pm0.9$
0.5 wt%	side	$1661 \pm 26$	$16.9\pm0.1$	$12.7\pm0.3$	$4.6\pm0.1$	$41.5\pm0.5$	$29.6 \pm 1.5$
1.0 wt%	hopper	$1654\pm48$	$16.4\pm0.1$	$12.6\pm0.4$	$5.6\pm0.2$	$28.8 \pm 1.5$	$23.9 \pm 1.2$
1.0 wt%	side	$1701 \pm 29$	$16.6\pm0.1$	$12.5\pm0.3$	$5.2\pm0.1$	$28.5\pm1.9$	$26.0\pm0.5$
2.0 wt%	hopper	$1578 \pm 52$	$16.8\pm0.1$	$13.5\pm0.3$	$4.9\pm0.2$	$20.2 \pm 1.1$	$28.9\pm2.2$
2.0 wt%	side	$1531 \pm 54$	$16.7\pm0.1$	$13.5\pm0.4$	$4.8\pm0.1$	$18.5\pm1.6$	$28.8\pm2.1$
5.0 wt%	hopper	$1604 \pm 102$	$17.0\pm0.3$	$14.5\pm0.4$	$4.5\pm0.1$	$11.7 \pm 1.3$	$23.4\pm1.0$
5.0 wt%	side	$1632\pm50$	$16.7\pm0.3$	$14.5\pm0.7$	$4.5\pm0.2$	$10.6\pm1.1$	$22.6\pm1.2$

Table 1. Influence of feeding condition of Nanocyl<sup>TM</sup> NC7000 on the mechanical properties\* of PP based composites

\*E<sub>t</sub> Young' modulus,  $\sigma_Y$  yield stress,  $\sigma_M$  maximum stress,  $\sigma_B$  stress at break,  $\epsilon_Y$  strain at yield point,  $\epsilon_M$  strain at maximum stress,  $\epsilon_B$  strain at break,  $a_{cA+23}$  notched Charpy impact strength at 23°C

Table 2. Influence of feeding condition of Baytubes<sup>®</sup> C150P on the mechanical properties of PP based composites \*

MWCNT	Feeding	E <sub>t</sub>	$\sigma_{\rm Y} = \sigma_{\rm M}$	σ <sub>B</sub>	$\varepsilon_{\rm Y} = \varepsilon_{\rm M}$ [%]	ε <sub>B</sub>	a <sub>cA+23</sub>
C150P	location	[MPa]	[MPa]	[MPa]		[%]	[kJ/m <sup>2</sup> ]
without 2.0 wt% 2.0 wt% 5.0 wt%	- hopper side hopper side	$\begin{array}{c} 1598 \pm 58 \\ 1593 \pm 61 \\ 1488 \pm 44 \\ 1582 \pm 38 \\ 1464 \pm 34 \end{array}$	$\begin{array}{c} 17.1 \pm 0.1 \\ 16.4 \pm 0.2 \\ 15.8 \pm 0.1 \\ 15.9 \pm 0.1 \\ 15.1 \pm 0.1 \end{array}$	$\begin{array}{c} 11.4 \pm 0.1 \\ 13.4 \pm 0.6 \\ 13.6 \pm 0.3 \\ 13.8 \pm 0.4 \\ 13.7 \pm 0.3 \end{array}$	$\begin{array}{c} 4.7 \pm 0.1 \\ 4.7 \pm 0.2 \\ 5.3 \pm 0.1 \\ 4.3 \pm 0.1 \\ 4.5 \pm 0.3 \end{array}$	$\begin{array}{c} 31.3 \pm 4.2 \\ 17.2 \pm 2.5 \\ 16.8 \pm 1.4 \\ 11.1 \pm 1.2 \\ 10.2 \pm 1.6 \end{array}$	$\begin{array}{c} 37.6 \pm 1.8 \\ 26.4 \pm 1.2 \\ 16.9 \pm 0.9 \\ 21.2 \pm 1.0 \\ 15.4 \pm 1.8 \end{array}$

\***E**<sub>t</sub> Young' modulus,  $\sigma_Y$  yield stress,  $\sigma_M$  maximum stress,  $\sigma_B$  stress at break,  $\epsilon_Y$  strain at yield point,  $\epsilon_M$  strain at maximum stress,  $\epsilon_B$  strain at break,  $\mathbf{a}_{cA+23}$  notched Charpy impact strength at 23°C

## Figure captions

**Figure 1** Schema of the injection molded plate (80 x 80 x 2 mm<sup>3</sup>) and measuring directions

**Figure 2.** Scanning electron microscopy images of pristine MWCNT as-received powders at two magnifications: (A) Nanocyl<sup>TM</sup> NC7000, (B) Baytubes<sup>®</sup> C150P

**Figure 3.** Electrical percolation behavior of Nanocyl<sup>™</sup> NC7000 and Baytubes<sup>®</sup> C150P in PP measured on compression molded samples (small-scale mixing, DSM15)

**Figure 4.** Influence of feeding condition of Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000 on the volume resistivity in compression molded PP composites (compounded using ZE25) at different nanotubes loadings (wt%)

**Figure 5.** Influence of feeding condition of Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000 on the electrical surface resistivity in compression molded PP composites (compounded using ZE25) at different nanotubes loadings (wt%)

**Figure 6** Influence of feeding position of Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000 on the electrical volume resistivity in injection molded PP composites (compounded using ZE25) at different nanotubes loadings (wt%)

**Figure 7** Influence of feeding position of Baytubes<sup>®</sup> C150P and Nanocyl<sup>TM</sup> NC7000 on the electrical surface resistivity in injection molded PP composites (compounded using ZE25) at different nanotubes loadings (wt%)

Figure 8 Nanotube agglomerate area ratio in polypropylene composites versus the contents of Nanocyl<sup>TM</sup> NC7000 or Baytubes<sup>®</sup> C150P

**Figure 9** Characteristic light microscopy images (section thickness  $10\mu$ m) illustrating the influence of feeding position on remaining primary agglomerates in PP composites including the agglomerate area fraction A/A<sub>0</sub>: (A) 2 wt% Nanocyl<sup>TM</sup> NC7000, (B) 2 wt% Baytubes<sup>®</sup> C150P

Figure 10 Stress-strain behavior of PP composites with Nanocyl<sup>TM</sup> NC7000

compounded using different feeding positions

**Figure 11** Stress-strain behavior of PP composites with Baytubes<sup>®</sup> C150P compounded using different feeding positions

Figures:







Figure 2.







Figure 4



Figure 5



Figure 6



Figure 7



Figure 8



Figure 9



Figure 10



Figure 11

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