

Multifller carbon nanotube, graphene, and carbon black composite flaments: A path to versatile electromaterials

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Addressing the growing demand for conductive and flexible composites, this research focuses on producing thermoplastic composite fibers made of polyurethane and carbon nanomaterials featuring the highest possible electrical conductivity. Based on a recently developed methodology enabling the formation of very high filler contents of 40% w/w, this work presents a systematic investigation of the role of all the materials used during the manufacturing process and selects the materials that ensure the best electrical performance. The results show that the highest electrical conductivity and current-carrying capacities are obtained when dimethylformamide is used as a solvent, and small amounts of AKM surfactant aid the de-agglomeration of carbon nanomaterials. It is also shown that the hybridization of MWCNTs fller with graphene nanoplatelets and small amounts of carbon black is beneficial for the electrical properties. However, the highest performance is achieved with SWCNTs as fllers, exhibiting two orders of magnitude higher electrical conductivities of 6.17 \times 10⁴ S/m.

Introduction

The quest for innovative materials with tailored properties has led to remarkable advancements in the field of composite materials in recent years.^{[1](#page-6-0)} These materials, formed by combining two or more distinct components, offer a wide spectrum of possibilities for designing materials with superior mechanical, electrical, and thermal properties. Among these composites, polymer-carbon nanomaterials composites have gained increasing attention due to their potential in many applications across diverse industries. $2-4$ $2-4$

Carbon nanomaterials, particularly carbon nanotubes and graphene, when used as fillers, offer a unique blend of properties, including very high electrical conductivity, thermal conductivity, mechanical robustness, lightweight, and corrosion resis t ance.^{[5–](#page-6-3)[9](#page-6-4)} Polymers, on the other hand, are also lightweight and corrosion-resistant materials. Additionally, they are inexpensive and easy to process, which, combined with their tunable mechanical properties, makes them a very popular material in countless applications both in daily life and industry. Incorporating carbon nanomaterial fillers into these polymers extends the range of their potential applications. Polymers enriched with carbon nanomaterials could become stronger, electrically and thermally

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The article presents a pioneering exploration into the synthesis and application of a novel composite material. This research significantly impacts the field of electromaterials by introducing a cutting-edge approach that leverages the synergistic properties of carbon nanotubes, graphene, and carbon black within a single flament. The impact of this research extends beyond the laboratory, infuencing the development of next-generation materials that bridge the gap between conventional materials and advanced nanomaterials. The presented composite filaments open avenues for the creation of innovative devices and systems that demand good mechanical strength, electrical conductivity, and thermal stability. Moreover, the versatility of these flaments allows for the optimization of materials properties, enabling customization based on specific application requirements. In addition to its technological significance, the paper contributes to sustainability efforts by facilitating the production of lightweight, energy-efficient materials. The insights provided by this research have the potential to reshape the landscape of materials science, inspiring further exploration and innovation in the quest for versatile and highperformance electromaterials.

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conductive while retaining their lightweight nature and, in many cases, processability.

Due to its amazing versatility, one of the very interesting polymers is thermoplastic polyurethane (TPU) .^{[10](#page-6-5),11} This block copolymer is composed of soft and rigid segments. By fne-tuning its chemical structure, TPU can have a range of mechanical properties, from soft and fexible elastomers to rigid plastics of high modulus. Additionally, TPU can be both melt and solution-processed. Therefore, it can be formed by a variety of processes, such as injection molding, extrusion, or three-dimensional (3D) printing, and be used for spray coating or impregnation.

As a result, TPU is used for a wide range of applications, including sporting goods, technical textiles, medical devices, automotive and aerospace equipment, packaging, and 3D printing flaments. Increasing the range of these applications by the formation of composites would be thus highly interesting and easily fnd many applications. For this reason, many studies have recently focused on the formation of composites formed by TPU and carbon nanomaterials to obtain electrically conductive materials with better mechanical, thermal, or fammability properties compared to pure TPU, $^{12-14}$ for sensing, 15,16 15,16 15,16 15,16 15,16 heating,¹² or EMI shielding^{12,[17](#page-6-11),18} applications. To address the variety of potential applications, the composites were prepared in many forms, including sheets, filaments, and foams, $19-21$ $19-21$ $19-21$ using various methods and fillers.^{[18](#page-6-12),[19,](#page-7-0)[22](#page-7-2),23} It is important to mention that the properties of specifc composites depend on many factors, including the chosen fller, processing method, or the composite form to be obtained.

Our recent study focused on manufacturing carbon nano-tube—TPU filaments.^{[20](#page-7-4)} Filaments are an interesting form of composites that enables their convenient use as sensors, base materials for thermal transfer, or 3D printing. One of the frst major areas of research concerning the use of carbon nanotubes was in the field of polymer composite fibers. $24-27$ $24-27$ Despite the fact that CNT-containing fbers can achieve high conductivity of up to 50 S/cm, the choice of polymers that could be used at that time was severely limited. For this reason, research has focused on producing fbers only from CNTs; however, due to their properties, these fibers have limited elas-ticity, which is crucial in the textile industry.^{[28](#page-7-7)} Targeting the high electrical conductivity of these composites, we developed a method that enabled the manufacturing of composites with a very high loading fraction of CNTs (40% w/w). Such loading fraction ensured obtaining an order of magnitude higher conductivity than reported so far for analogous composites, $29-32$ $29-32$ $29-32$ where composites were obtained with conductivities of: 26.3 S/m, 116.98 S/m, 677 S/m, and less than 10^{-3} S/m, respectively.

The manufacturing process is very important for optimizing the electrical properties of the composites; however, it is the fller which is the key to obtaining electric current percolation pathways within the insulating polymeric matrix. In this case, the composite's conductivity depends on many aspects, which can be generally divided into two categories of intrinsic and

extrinsic factors. Intrinsic factors will include the conductivity of micro-/nanoparticles originating from their elemental composition and atomic arrangement (e.g., graphene with pure sp^2 bonding will be more conductive than amorphous carbon). 33 This category will also include structure-related bandgap size, concentration of defects, type of defects or doping type, and level. On the other hand, extrinsic factors will include the concentration (weight percent) of the nano-/microparticles, which should be as high as possible to form electric current percolation pathways efectively, but should not compromise the desired mechanical properties. Further factors are the size, shape, and aspect ratio of the particles.^{[34,](#page-7-11)[35](#page-7-12)} For example, we may expect that large graphene platelets or higher aspect ratio carbon nanotubes will facilitate the formation of the percolation pathways. It has also been shown in the literature that mixing diferent sizes and shapes of nano-/microparticles may increase conductivity. 36 The larger particles may work like bridges, while the smaller ones may become the fllers of voids. In the case of such particles as carbon nanotubes, conductivity also depends strongly on alignment due to the high anisotropy of its properties.^{[37](#page-7-14)}

Finally, it is important to mention processing-related factors—agglomeration and uniformity of distribution of nanoand microparticles, which, if not appropriately addressed, do not ensure good electrical performance.

In the following, we test the possibility of increasing the conductivity further via the change in processing methodology and fller composition. We, therefore, examine the new capabilities of the developed method for producing highly conductive composites. We show that the hybrid carbon composites, with sufficient fabrication processes enabling their dense packing, outperform the parameters of previously known similar heterophase materials. In the paper, we also undertake the verifcation whether the change of components such as solvent or surfactant afects the parameters of the fnal composite.

Materials and methods

Materials

The functional phase of the composites was prepared based on NC7000 multiwalled carbon nanotubes (MWCNTs) from Nanocyl SA (Sambreville, Belgium). Additionally, two different types of graphenes were employed. The first type, denoted as G1, comprised nanoplatelets obtained from Cheap-Tube (Cambridgeport, Vt., USA), characterized by a thickness ranging from 8 to 15 nm and a diameter of 1–2 μm. The second type, G2, was produced by XG Sciences (Lansing, Mich., USA) and exhibited a 6- to 8-nm thickness with a diameter of 5 μm, per the provided datasheet. Further materials used as functional phase included carbon black (CB) from Graphene Supermarket (New York, USA) with a particle size of 30 nm and a surface area of 254 m^2/g as well as highly conductive single-walled carbon nanotubes (SWCNTs) purchased from Sigma Aldrich (Darmstadt, Germany), with ≥93% carbon

content, SWNT length of >5 µm, and diameter in the range of 1.2–2.0 nm.

The composite matrix was made of thermoplastic polyurethane (TPU) purchased from BASF (Ludwigshafen, Germany) with a catalogue name Elastollan soft 35AP. According to the datasheet, its density amounts to 1.18×10^3 kg/m³.

In this study, two diferent solvents were employed. Both tetrahydrofuran (THF) with a molecular weight of 72.11 g/ mol and *N*,*N*-dimethylformamide (DMF) with a molecular weight of 73.09 g/mol were purchased from Sigma Aldrich (Darmstadt, Germany). The surfactants tested included AKM - 0531 (AKM) from NOF Corporation (Tokyo, Japan) and Triton X-100 (Triton) purchased from Sigma Aldrich (Darmstadt, Germany).

Manufacturing process

Composite preparation

The composites were prepared following the previously described method.[20](#page-7-4) Initially, all components of the functional phase were accurately weighed and combined in appropriate ratios along with the surfactant. The solvent was then added in a sufficient quantity, allowing the particles to de-agglomerate freely during the sonication process. Deagglomeration was carried out using a VCX 750 ultrasonic homogenizer with the amplitude set at 80% of nominal power for 30 min. In the next step, the weighed amount of polymer was added. The composite was stirred on a Heidolph MR - Hei magnetic stirrer at several Celsius degrees below the solvent's boiling point until the solvent was preliminarily evaporated.

Next, the composites were transferred to a Czylok FCC 7-SM laboratory oven for complete solvent evaporation. The drying process was conducted in two steps to ensure thorough solvent evaporation without rapid drying. The working temperatures for both steps were selected based on the specifc solvent used (50°C and 80°C for THF and 100°C and 150°C for DMF). Each drying step lasted 5 h.

Extrusion process

After the composite material was cooled and cut into pellets, it was placed in a purpose-designed screw extruder.^{[20](#page-7-4)} This extruder facilitates the formation of flaments from the composite material. The machine has two heating zones: the frst set at 170°C and the second at 190°C. The screw works at a constant speed of 38 rpm. The machine allows the extrusion of flaments of diferent diameters, adjustable by the nozzle. In this work, a nozzle with a diameter of 1.8 mm was used.

Testing of the composite flaments

The resistance of the flament samples was measured by the two-point method. The samples were coated with room-temperature drying silver-paint Electrolube, Leicestershire, UK. Each measurement was performed on five 60-mm-long samples. The measurements were performed using True

RMS UT804 multimeter, Uni-Trend Technology Co., Ltd., Guangdong, China.

The current-carrying capacity was measured in a purpose-built setup composed of a QL564P DC power supply, Aim TTi, Huntingdon, UK, a Keithley 2000 DC multimeter, Keithley Instruments, Cleveland, Ohio, USA, both controlled by dedicated LabVIEW software, National Instruments Corporation, Austin, Texas, USA. The current was increased at a rate of 10 mA/s up to 2A or thermal failure of the composite.

For the investigation of electrical properties, two parameters were considered, namely absolute electrical conductivity (σ) and specific conductivity (σ') often used to characterize CNT-based materials. 28 These parameters were calculated using the following formulas:

$$
\sigma = \frac{1}{R} \frac{L}{A} \left[\frac{m}{\Omega \cdot m^2} = \frac{S}{m} \right] \text{ and } \sigma' = \frac{1}{R} \frac{L^2}{m} \left[\frac{S \cdot m^2}{kg} \right]
$$

where *R* is the resistance in Ω , *L* is the length in m, *A* is the cross-sectional area in m^2 , and *m* is the mass of the sample in kg.

The static tensile test was performed on 40-mm-long fibers, with a test section length of 20 mm and 10 mm clamped in the jaws on each side, on a QC-506B1 material testing machine manufactured by Cometech Testing Machines Co., Ltd, Taichung City, Taiwan.

Results and discussion

The infuence of surfactants and solvents on the electrical performance of the composites

The frst part of the study focused on the manufacturing process and investigated the infuence of surfactants and solvents on the electrical performance of the composites. The composites with the highest MWCNT concentration of 40% w/w developed in our previous study were chosen as the base material. 20

First, the change of surfactant was studied. The tested surfactants included Triton and AKM. The experiments followed the protocols developed previously.²⁰ In brief, MWCNTs with a concentration of at most 2% were sonicated in THF. Next, 2 wt% of a chosen surfactant in relation to MWCNTs was added and sonicated further. Finally, TPU was mixed into the dispersion. The dispersion was then dried and extruded in the form of a flament.

The obtained filaments were tested electrically. The obtained absolute conductivity values were $1.28 \cdot 10^3$ S/m for AKM and $3.89 \cdot 10^2$ S/m for Triton, and the specific conductivity values were $9.76 \cdot 10^{-1}$ S m²/kg and $2.81 \cdot 10^{-1}$ S m²/kg, respectively.

The results indicate that selecting an unsuitable surfactant can signifcantly degrade the material's conductivity. The sample containing AKM showed an order of magnitude better conductivity. Therefore, further research was limited to the AKM surfactant.

Next, tests investigated the effect of using different amounts of AKM and diferent solvents. Both the amount of surfactant and type of solvent can infuence the properties of the manufactured composite. However, the accurate amount of surfactant could also be dependent on the solvent type, as the solubility of the surfactant is solvent-dependent. The choice of solvents was limited by the solubility of the thermoplastic polymer Ellastolan soft 35AP and restricted to two solvents, specifcally DMF and THF. The results of the electrical parameters obtained are shown in **Table** [I.](#page-3-0)

Analysis of the results presented in Table [I](#page-3-0) shows that using 10% AKM signifcantly decreases the electrical conductivity of the composites independent of the solvent used. This could be explained by the fact that surfactants are in a nonconductive phase. They have a crucial role in separating CNTs and inhibiting their agglomeration in the composite, which improves the percolation of electric current through the composite. However, with larger amounts of surfactants, a decrease in the electrical conductivity of the composite is observed. That could be caused by the amphiphilic agents, added in excess, not only homogenize the composite by reducing the agglomerates of carbon particles, but at the same time they may isolate some CNT nanoparticles through their presence, which inhibits the transportation of charge between them.

The results presented in Table [I](#page-3-0) also showed that when DMF was used as the solvent, the electrical conductivities of the fnal composite were higher. This means that the choice

of solvent signifcantly afects the dissolution of the polymer and its bonding to the functional phase, which can improve the properties of the fnal composite.

The electrical testing of composite filaments prepared using diferent solvents and diferent amounts of surfactant also included an investigation of their current-carrying capacity. The results are presented in **Figure** [1b](#page-3-1), along with a photo of an example specimen of the tested composites (Figure [1](#page-3-1)a).

The obtained results follow the previous observations and discussion. First, increasing the AKM amount from 2% to 10% deteriorates the electrical performance of the flaments independent of the solvent used. For the composite prepared using THF solvent, there was a decrease in the maximum current recorded by 0.35 A and for those containing DMF by 0.2 A. However, the choice of the solvent itself, with the same amount of surfactant for both 2% and 10%, also signifcantly afected the value of the maximum current recorded. The current-carrying capacity of DMF-based flament was higher than those for which THF was used (diference of 0.15 A for 2% and 0.3 A for 10% samples).

Analysis of the previously discussed results showed that the best electrical performance of the composite flaments is obtained when DMF is used as solvent and AKM as surfactant. The 2% of AKM in relation to the MWCNT weight allows de-agglomeration of the carbon particles, which is clearly visible during composite preparation, as a clear foaming of the sonicated carbon particles is observed. This content, therefore, allowed dense packing of the functional phase, which translated into high electrical conductivity of the composite. We considered that any higher amount of surfactant, even if it caused an increase in homogenization would result in a decrease in electrical parameters; hence, this content was chosen for further studies. Therefore, this manufacturing protocol was chosen for the following experiments.

Functional phase modifcation

As explained in the introduction, although the manufacturing process is very important for optimizing the electrical properties

> of composites, the fller, as an electrically conductive phase, is the key determinant afecting the electrical properties. Considering this, we first tested the possibility of enhancing the electrical performance of the composite flaments by hybridizing MWCNTs with other carbon nanomaterials (i.e., nanomaterials of diferent atomic structures, sizes, and shapes). The latter fllers included two types of commercial graphene

nanoplatelets—G1, characterized by a thickness of 8–15 nm and diameter of 1–2 mm; G2 of 6–8 nm thickness and 5 mm in diameter, as well as commercial carbon black with a particle size of 30 nm and a surface area of 254 m²/g.

The scanning electron microscope images of the chosen fllers are presented in **Figure** [2a](#page-4-0)–c. The images clearly illustrate the diferences in the structures of these materials. The clear forms of MWCNT branches (Figure [2](#page-4-0)a), the fake structure of graphene powder (Figure [2](#page-4-0)b), and the spherical form of carbon black (Figure [2](#page-4-0)c) are evident. The diferences in the dimensions of the structures are also apparent, consistent with the manufacturers' declarations in the materials' datasheets.

The fller mixtures were composed of either MWCNTsgraphene nanoplatelets or MWCNTS-carbon black in two ratios of 1:1 and 3:1. The total fller ratio in every composite amounted to 40% w/w. Composites with 30% G1/G2 and 10% MWCNT content were not considered in the work, as such a phase ratio resulted in a composite being overly brittle. The manufacturing procedure followed the previously proposed procedure based on sonication and evaporation of the excess solvent during mixing, which should guarantee effective de-agglomeration of the functional phase particles and their uniform distribution in the composite. However, modifying the functional phase's shapes and dimensions can change the structure at the nanoscale. For verifcation of whether hybridization with either fake graphene or spherical carbon black structure will change the homogeneity of the cross-sectional structure of the flament, we performed SEM image analysis for composites flled purely with MWCNTs as well as MWCNTs-graphene G1 and CNT-carbon black hybrid composites for a 1:1 ratio (Figure [2d](#page-4-0)–f).

Diferent flament morphologies can be observed in the presented images due to the apparent presence of diferent forms of nanocarbons in the composites. The cross-sectional images indicate a uniform distribution of diferent carbon nanostructures in the composite fbers, both in the case of pure MWCNT fller (Figure [2](#page-4-0)d) and MWCNTs-graphene (Figure [2e](#page-4-0)) and MWCNT-carbon black (Figure [2](#page-4-0)f) fllers.

All prepared samples were tested electrically. As shown in **Table** [II](#page-5-0), in the case of MWCNT-graphene nanoplatelet samples, both absolute and specifc conductivity increased compared to samples containing only MWCNTs. In the case of samples containing 30% w/w of MWCNTs and 10% of G1, the absolute and specifc conductivity increased by 17% and 10%, respectively. For the 1:1 ratio of MWCNTs and G1, the increase was higher and amounted to 37% and 48% for absolute and specifc conductivity, respectively. This indicates that large G1 nanoplatelets of 1–2 mm contribute to better formation of percolation pathways. Larger G2 nanoplatelets (5 mm in diameter) are even more effective in forming current pathways. In the case of MWCNTs-G2 samples, the absolute conductivity increased by 10% for the 3:1 ratio and 106% for the 1:1 ratio, while specifc conductivity increased by 15% and 120% for the 3:1 and 1:1 ratio, respectively. In the case of carbon black, the efect is quite diferent. These amorphous carbon particles of 30 nm in diameter improve the conductivity (absolute by 51% and specifc by 49%) when added in small amounts (3:1 ratio) by flling the gaps between high aspect ratio carbon nanotubes. However, an increase in their share to 20% w/w (1:1 ratio) is no longer beneficial and results in an apparent decrease in conductivity of −25% for absolute conductivity and −26% for specifc conductivity. It is worth mentioning that these results are an order of magnitude higher than for TPU-CNT composites produced by other methods that do not allow such dense packing of the functional phase.^{38-[40](#page-7-16)}

Figure 2. Scanning electron microscope images of nanocarbon structures (a) multiwalled carbon nanotubes (MWCNTs); (b) graphene nanoplatelets G1; and (c) carbon black; used in hybrid fibers together with the images of fber composites containing those nanocarbons (d) 40% MWCNTs; (e) 20%CNT20%G1; (f) 20%CNT20%CB.

Some more insight into the effect of the hybridization of fillers on the electrical properties of the flaments has been obtained from the current-carrying capacity tests presented in **Figure** [3](#page-5-1). The results showed that the fibers with 10% graphene content of both G1 and G2 could not carry the same current as with carbon nanotubes alone (a decrease of 0.11 A (−7%) and 0.13 A (−8%), respectively). However, at 20% graphene content, both composites reached higher current values (an increase of 0.19 A (12%)

Table II. Absolute and specific conductivities for fibers of varying

and 0.36 A (23%), respectively). This result indicates that the large graphene platelets may be less efective than CNTs in heat removal. However, a signifcant improvement in the formation of conductive pathways for a 1:1 ratio will help overcome this issue. In the case of carbon black, the results are in good agreement with the conductivity measurements and conclusions. The 3:1 ratio of CB caused a slight increase in current-carrying capacity thanks to denser phase packing in the composite, resulting in better electrical connections. However, at a 1:1 ratio, replacing the carbon nanotubes with an excessive amount of carbon black resulted in a reduction of maximum current.

Considering all the filler parameters that may affect the electrical transport within the composite, one more fller has been tested. The industrial-grade MWCNTs have been replaced by single-walled CNTs, which are expected to be less defective and thus more conductive intrinsically than industrial-grade MWCNTs. Like all previous samples, the SWCNT composite contained 40 weight percent of the fller. The SWCNT indeed showed much better electrical performance than all previous samples. The absolute conductivity (Table [II](#page-5-0)) was two orders of magnitude higher than the composite containing MWCNTs. The specifc conductivity was one order of magnitude higher for SWCNTs than for MWCNTs. The composite based on SWCNTs carbon nanotubes showed the ability to carry a signifcantly higher current (Figure [3](#page-5-1)). The value obtained was almost 2.5 times higher than for MWCNTs. This means that the proper selection of CNTs contained in the functional phase can drastically improve the properties of the composite.

In the further step, the mechanical properties of fbers were compared to verify the infuence of modifcation of the functional phase. The exact results are shown in **Table** [III](#page-6-13).

The results indicate that replacing the industrial-grade, multiwalled nanotubes with high-quality single-walled ones caused the material to be able to operate at signifcantly higher stresses (an increase of almost 3 MPa). Hybridizing MWCNTs with graphene, in both ratios tested, resulted in a reduction of the maximum stresses, inducing specimen breakage. As for the material's tensile susceptibility, both with SWCNT or graphene,

the recorded changes were marginal (not exceeding a few percentages). For the hybridization of carbon nanotubes with carbon black, the average maximum strains were similar to those recorded for the composite containing the MWCNTs solely.

Conclusions

This article investigated the role of materials selection used in the manufacturing process on the electrical performance of the TPU-carbon nanomaterials composite flaments. The analysis shows that every substance used during the manufacturing process may affect the final electrical properties of the flament.

The modifcations of the manufacturing process of TPU flament with 40% w/w of MWCNTs showed that a properly selected surfactant can significantly improve the electrical properties. Simply changing the surfactant used from Triton to AKM in the fber preparation process resulted in almost three times higher conductivity (increase from $3.89 \cdot 10^2$ S/m to $1.28 \cdot 10^3$ S/m). The material produced with AKM also showed the ability to operate at a higher maximum current. The research also showed that the amount of surfactant used also plays a key role. For both the composites prepared with THF and DMF, we observed that increasing the amount of surfactant from 2% to 10% deteriorated the electrical properties of the material.

The solvent used during fabrication, although being evaporated and therefore absent in the fnal flament, also appears to afect the properties of the composite. Composites containing DMF had better electrical conductivity than those with THF for both composites containing 2% and 10% of the surfactant and were capable of conducting a higher current.

Finally, it is crucial to choose the proper functional phase. The results showed that when cheaper and, therefore, more achievable multiwalled nanotubes need to be used, it is benefcial to hybridize them with other materials. Addition of graphene nanoplatelets can signifcantly improve the electrical properties as in the case of 20% of graphene G2 where the obtained conductivity was almost twice as high even though the maximum stress at which the fber could work had decreased. Analysis of the results obtained also showed that the use of a small amount of carbon black can improve the conductivity of the material. However, in the case of this material, the chosen amount must be optimized because, by replacing the carbon nanotubes too excessively with carbon black, the electrical properties of the material were lowered relative to the original composite.

However, if less restricted by the price of the composite, the SWCNTs are a much better choice, ensuring two orders of magnitude higher absolute electrical conductivity, an order of magnitude higher specifc conductivity, and 2.5 times higher current-carrying capacity. Fibers based on SWCNTs showed higher maximum stress resistance.

As mentioned in previous work, 20 CNT-TPU fiber composites can be used in many applications such as smart clothing, textiles, or 3D printing. The improvement in electrical properties we obtained in this work brings us closer to this destination.

Author contributions

Conceptualization, S.L.K, A.L.R.; methodology, S.L.K.; validation and formal analysis, S.L.K.; investigation, L.N., S.L.K; writing—original draft preparation, L.N., A.L.R.; writing review and editing, S.L.K.; discussion of results A.L.R, M.J; supervision, S.L.K, M.J.

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Data availability

Not applicable.

Conflict of interest

The authors declare no confict of interest.

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