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**Stretchable Conductive Fibers of Ultra-high Tensile Strain and
Stable Conductance Enabled by Worm-shape Graphene Microlayer**

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Abstract

Stretchable electrical conductors have demonstrated promising potentials in a wide
range of wearable electronic devices, the conductivity of most of reported stretchable
conductive fibers will be changed if be stretched or strained. But however, stable

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1 conductance is essential for wearable and stretchable device, to ensure the
2 performance of wearable devices is stable. Inspired by the peristaltic behavior of
3 arthropods, we designed a graphene coating similar to the caterpillar structure on the
4 PU fiber surface, enabled by coating the worm-shape graphene microlayer onto
5 polyurethane filaments. Such worm-shape filaments can be stretched up to 1010%
6 with wide reversible electro-response range ($0 < \epsilon < 815\%$), long-term durability (>4000
7 stretching/releasing cycles), good initial conductivity ($\sigma_0 = 124 \text{ S m}^{-1}$) and high-quality
8 factor ($Q = 11.26$). Remarkably, the worm-shape filaments show distinctive
9 strain-insensitive behavior ($\Delta R/R_0 < 0.1$) up to 220% strain. Furthermore, the filaments
10 as electrical circuits of LED to track signals from robust human joint movements are
11 also demonstrated for practical application. Such worm-shape filaments with
12 distinctive strain-insensitive behavior provide a direct pathway for stretchy
13 electronics.

14 **Keywords:** *graphene, bionic, strain-insensitive, polyurethane filaments, stretchy*
15 *electronic.*

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1 Smart wearable device, as an important case of artificial intelligence, is a
2 complex micro-system composed of electronic skin,¹⁻³ actuators,⁴⁻⁶ stretchable
3 electronic circuits,⁷⁻⁹ flexible supercapacitors¹⁰⁻¹² and strain sensors¹³⁻¹⁵ *etc.* The
4 conductivity of most of reported stretchable conductive fibers will changed if be
5 stretched or strained, and these conductive fibers could be suitable as sensors. But
6 stable conductance is essential for wearable and stretchable device, to ensure the
7 performance of wearable devices is stable. Electronic circuit is one kind of essential
8 electronic element to transfer electrical signal, which usually receives less awareness
9 and attentions for wearable devices. However, under various robust body movements,
10 commercial metal electronic circuits might affect or even change the electrical signal
11 of smart clothes and wearable devices.¹⁶⁻¹⁸ Besides, such rigid circuits could also
12 restrict limb movements and unsatisfied wearing experience. Therefore, stretchable
13 electronic circuits with high stretchability and strain-insensitivity are the desirable
14 electronic element for wearable devices and smart clothes.

15 A growing number of strain-insensitive stretchy electronics are reported for smart
16 wearable devices. Currently, two-dimensional micro or nano films with invariant
17 resistant changes are the mostly proposed strategies for stretchy electric circuits such
18 as electrospun elastomeric mats decorated with silver nanoparticles,¹⁹ self-generated
19 microfibril network on the SBS film,²⁰ the fractal gold nanomesh-like structure,²¹ and
20 stretchable wirings on Elastic substrate.²² However, these 2D stretchy electronics with
21 various micro-patterns could only possess negligible resistant changes under
22 low-level strains (<100%), which is not suitable for tracking robust human joint

1 movement (strains>100%). 1D fiber-shaped electronics might be the ideal structure
2 with high stretchability, Baughman *et al.* have reported 1D microlayer sheath-core
3 conducting fibers with microlayer buckling,²³ enabling a resistance change of less
4 than 5% for a 1000% stretch of the rubber fibers, but such rubber fibers (ropes) with a
5 large diameter 1.7 mm might restrict the applications for smart clothes. Therefore, the
6 criterion for ideal fiber-shaped electronics generally includes small diameter,
7 strain-insensitivity under high-level strain and good durability.

8 Herein, inspired from the creeping of worms with an effective and logical
9 deformation, we fabricated the worm-shape filaments by coating worm-shape
10 graphene microlayer onto polyurethane filaments. Such bionic micro-structures
11 exhibit distinctive strain-insensitive behavior $\Delta R/R_0$ smaller than 0.1 up to 220%
12 stretch, along with long-term durability after 4000 stretching/releasing cycles, good
13 initial conductivity of 124 S m⁻¹ and high-quality factor ($Q=11.26$). Compared with
14 other stretchable electronics, inspired by the peristaltic behavior of arthropods, we
15 designed a graphene coating similar to the caterpillar structure on the PU filament
16 surface. When stretched, our design is similar to the creep behavior, maintaining a
17 stable electrical conductivity. The as-obtained worm-shape graphene microlayer PU
18 filaments exhibit ultra-high stretchable, excellent strain-insensitive and more suitable
19 for wearable with better softness and comfort. Our worm-shape graphene microlayer
20 polyurethane filaments open an avenue for the fabrication of seamless connection
21 smart clothes and stretchable electronics.

22 RESULTS AND DISCUSSION

The processes for preparing of conductive polyurethane filaments with worm-shape graphene microlayer are illustrated in Figure 1a. The graphene/PU (GP) microlayer could be in-situ deposited on the surface of pristine PU filaments with the effect of non-solvent-induced phase separation.²⁴ GP_x@filament denotes that GP microlayer is deposited on a PU multifilament core under $x\%$ pre-stretching condition, which is detailed in the experimental section. In brief, pristine PU filament was stretched under the fabrication strain $x\%$, and then immersed into the graphene/PU inks for capillary absorption, afterwards, the GP microlayer could be in-situ formed on the soaked PU filaments in the coagulation water bath. After releasing the pre-stretching forces loaded on the filament substrate, the worm-shape graphene microlayer was achieved as the inset SEM of Figure 1a.

The diagram of the worm wriggle as in Figure 1b indicates the prototypical model of the GP_x@filament. The as-prepared GP_x@filament exhibits remarkable elastic, which can be stretched over 800% (GP₃₀₀@filament). Furthermore, GP₃₀₀@filament also exhibits strain-insensitivity as the stretchable electric circuits in Figure 1c, the luminance of LEDs can be scarcely changed with the effect of robust elongation of elastic circuits. The evolution trend of the GP microlayer on GP₃₀₀@filament are shown in Figure 1d under strains from 0% to 250%, the peak width tends to elongate while the peak height decrease owing to the flattening trend under the strain, and the peak unit would quickly recover to the original shape once releasing the strain. The structure of GP microlayer maintains well and the LED is always on even under 250% strain indicating the well strain-insensitive behavior under robust elongation.

Figure 1. Bionic superelastic conductive polyurethane filaments with worm-shape graphene microlayer. (a) Schematic diagram of preparing the worm-shape filaments. (b) Prototypical model and photographs of a GP₃₀₀@filament pull by 200 grams of weight. (c) LED patterned lights response with different elongation of GP₃₀₀@filament circuits. (d) The optical images of GP microlayer of GP₃₀₀@filament under stretching.

The morphology of GP₀@filament and GP₃₀₀@filament along with the pristine PU filament are illustrated in Figure 2a-f. From the longitudinal view of the filament, the bundle structure of GP₀@filaments (Figure 2c) is entirely covered by the GP microlayer, the increased diameter of GP₀@filament ($d=875\pm13\text{ }\mu\text{m}$) compared with the pristine PU filament ($d=840\pm10\text{ }\mu\text{m}$) implies the successfully deposited graphene microlayer. While the diameter of GP₃₀₀@filament ($d=775\pm22\text{ }\mu\text{m}$) tends to be smaller due to the pre-stretching treatment and the contraction force from the GP layer. Under the higher magnification, the worm-shape graphene microlayer can be obviously observed on the surface of GP₃₀₀@filament while the GP₀@filament with

1 smooth surface appearance. Furthermore, the graphene nanosheets are fixed stable on
2 the PU matrix from the inset of Figure 2c and Figure 2e. From the cross-section view
3 of the PU filament, the worm-shape graphene microlayer of GP₃₀₀@filament exhibit a
4 thicker microlayer (thickness: 107±11 μm) than the smooth graphene microlayer of
5 GP₀@filament (thickness: 74±7 μm), and the GP microlayer is entirely deposited on
6 the surface of PU filament without obviously gap or pore indicating the well interface
7 interaction between the micro-layer and PU substrate.

8 In addition, the FTIR spectra of the as-obtained graphene, PU, and
9 GP₃₀₀@filaments are depicted in Figure 2g, only -OH group and the benzenoid C=C
10 stretching band appeared on graphene without any oxygen groups. In contrast, the
11 carboxyl absorption peaks and C-O-C and C-H stretching bands of PU become
12 weaker indicating the sufficient blend of graphene and PU.²⁵ The graphene was
13 obtained by lab-scale high-pressure homogenizing method and Figure 2h shows the
14 orientation and exfoliation process preparing few-layer graphene in the flow path
15 during homogenizing process. The TEM images of the obtained graphene are
16 illustrated in Figure 2i. As shown in Figure 2i, the lateral size of the as-obtained
17 graphene is around 5~10 μm and the graphene with few-layers can be clearly
18 observed.

Figure 2. Morphology of the modified PU filaments, (a-f) SEM images of the pristine PU multifilament from longitudinal view (a) and cross section (b), GP_0 @filament (c) and (d), and GP_{300} @filament (e) and (f). (g) FTIR spectra of graphene, PU, and GP_{300} @filaments. (h) Orientation and exfoliation process of lab-scale pressure homogenizing method preparing few-layer graphene in the flow path during homogenizing process. (i) TEM image of as-obtain few-layer graphene.

We further evaluate the mechanical and electrical properties of such filaments. As shown in Figure 3a, all the samples exhibit remarkable superelastic characteristic, the elongation at break of the GP_x @filament is gradually decreased with the increasing $x\%$ owing to the residual internal stress arising from the pre-stretching treatment. For example, the elongation of GP_0 @filament is up to 1778% and GP_{300} @filament is

1 1010%. All the samples show almost the same breaking strength (~ 30 MPa) with low
2 modulus appearance. There are some random noise peaks on the stress-strain curve,
3 which resulted from the sudden breaking of the inside single PU filament.

4 The electrical conductivity curves of the stretchable $\text{GP}_x\text{@filaments}$ are
5 illustrated in Figure 3b. All the samples exhibit distinctly different electrical responses
6 with increasing axial stretching process. For instance, $\text{GP}_{300}\text{@filament}$ exhibits the
7 widest reversible electroresponse range up to 815% while $\text{GP}_0\text{@filament}$ could only
8 reach 50%, therefore, the wide reversibility electroresponse range is the intrinsic merit
9 of the stretchy wearable electronics. Thereinto, the conductivity of $\text{GP}_{300}\text{@filament}$
10 increases to 363 S m^{-1} at $\varepsilon = 300\%$ from 124 S m^{-1} at the free state ($\varepsilon = 0\%$), and then
11 quickly decreases to 28 S m^{-1} at $\varepsilon = 800\%$.

12 The variation of the initial electrical conductivity of the filament under 50%
13 stretching cycling process is shown in Figure 3c, nearly all the $\text{GP}_x\text{@filaments}$ exhibit
14 steady appearance with the robust stretching cycles except $\text{GP}_0\text{@filaments}$.
15 Furthermore, the strain dependence of relative resistance change $\Delta R/R_0$ is also
16 evaluate (Figure 3d), for all the $\text{GP}_x\text{@filaments}$, relative resistance changes increase
17 monotonically with increasing stretching process, and exhibit obviously lower rising
18 rate with increasing pre-stretching $x\%$. Choosing $\Delta R/R_0=0.1$ as the baseline,
19 $\text{GP}_0\text{@filaments}$ could quickly reach the baseline at 5% while $\text{GP}_{300}\text{@filaments}$ at
20 220% indicating the outstanding strain-insensitive performance.

21 Herein, the quality factor Q is employed to evaluate the electro-response of our
22 filaments under various strain conditions,²³ which is defined as the percent strain

divided by the percent resistance change. Compared with previous reported conducting coatings on elastomeric fibers or sheets,^{7, 8, 26-31} All the $\text{GP}_x\text{@filaments}$ exhibit acceptable Q level as shown in Figure 3e. The value of Q gradually increases from 4.62 of $\text{GP}_{50}\text{@filaments}$ to 11.26 of $\text{GP}_{300}\text{@filaments}$, which are all higher than the value of Q in the literature, indicating the outstanding strain-insensitive behaviors of the worm-shaped electronic circuits. Furthermore, in order to show the distinct function of the worm-shaped microlayer, a simple trial is carried out to monitor relative resistance changes of $\text{GP}_{300}\text{@filament}$ and $\text{GP}_0\text{@filament}$ under periodic loading and releasing process with various strain ratios (Figure 3f). It can be found that the relative resistance change of $\text{GP}_{300}\text{@filament}$ can be nearly negligible even at 300% strain (red line), in contrast, that of $\text{GP}_0\text{@filament}$ is more sensitive and the response range is within 50% strain (blue line). Such results reveal that pre-stretching treatment is the critical treatment for the stretchy electronics with the strain-insensitive properties.

Figure 3. Mechanical and electrical properties of such filaments. (a) Conductivity-strain

1 curves for $GP_x@$ filaments. (b) Conductivity-strain curves for $GP_x@$ filaments and theoretical
 2 curves for $GP_{300}@$ filament. (c) Variation of initial conductivity per centimeter during
 3 stretching-releasing cycles with 50% strain. (d) Relative resistance curves of $GP_x@$ filaments.
 4 (e) Comparison of quality factor Q for the $GP_x@$ filaments with data in literature. (f) Dynamic
 5 relative resistance of $GP_0@$ filament and $GP_{300}@$ filament under different strains with 2s cycle.

6
 7 In order to further clarify the mechanism of electrical conductive of worm-shaped
 8 graphene microlayer, the micro geometrical model of GP microlayer unit on
 9 $GP_x@$ filament during the stretching process is illustrated in Figure 4. The stretching
 10 process is classified into three simple stages, which can be divided by two boundary
 11 points. The contact critical point ($\varepsilon=\varepsilon_c$) is the special boundary that the two legs of the
 12 GP layer loop are point-contact, and the other boundary is the straighten
 13 pre-stretching state ($\varepsilon=x$) that the strain degree matches with the pre-stretching state.
 14 Therefore, the tendency of the micro-structure of GP microlayer generally passes
 15 through three phases separated by these two boundary points: the original state to the
 16 contact critical point ($0<\varepsilon\leq\varepsilon_c$), the contact critical point to the straighten pre-stretching
 17 state ($\varepsilon_c<\varepsilon\leq x$), and after the pre-stretching state ($\varepsilon>x$). The total resistance is expressed
 18 by an equation set which derived by Supplementary Method 1 and Supplementary
 19 Method 2. Based on this model, the initial value of A , D_0 and l of $GP_x@$ filament can
 20 be observed and calculated from Figure 2e, f. R_l and x are the variables which can be
 21 measured during the pre-stretching process. The free parameter (σ_d) is monotonously
 22 decreased to zero at $\varepsilon=\varepsilon_c$. The electrical conductivity of $GP_{300}@$ filament in Figure 3b

could almost match such model, however, the total calculated conductivity is slightly bigger than the measured conductivity, in the theoretical simulation, the hypothesis of this model was PU multi-filaments as uniform cylinder and the graphene microlayer as regular wrinkle layer. Actually, PU multi-filaments is consisted of single filaments with unordered structure leading to complex deformation, so the mechanical performance might be changed during stretching. In addition, the worm-shape graphene microlayer is randomly generated by the inter stress of microlayer, so the resultant wrinkle microlayer might be not so regular. Therefore, the discrepancy could occurred between theoretical and experimental results, but such discrepancy is within a reasonable margin of error.

Figure 4. Geometrical model of GP microlayer unit on GP_x@filament and the corresponding theoretical formula at different stages during the stretching process.

Figure 5 explores the mechanical hysteresis and strain-insensitive properties of GP₃₀₀@filament as example. GP₃₀₀@filament exhibits slightly mechanical hysteresis

1 and fine elastic recovery under different stretching cases from 20% to 140% in Figure
2 5a, indicating reliable resiliency. Figure 5b shows the reversible relative resistance
3 change of GP₃₀₀@filament under the wide strain range 0~400%, $\Delta R/R_0$ of the filament
4 is still under 0.8 even be elongated to 400% indicating the outstanding invariant
5 resistance changes under high level strain, and the hysteresis of the filament is as
6 small as 0.02 indicating the remarkable repeatability of GP₃₀₀@filament. The inset
7 exhibits tiny relative resistance change of 0.08 from 50% to 210% strain, which
8 reveals excellent stain-insensitive. Furthermore, the I-V curves is employed to
9 evaluate the electroresponse of the filaments under different stretching strain levels
10 (from 0 to 300%). Under 5 V on-load voltage, the I-V curves (Figure 5c) of the
11 filaments exhibit linear shape indicating the intrinsic properties of good conductor,
12 and the slope of these curves only decrease 21% for 300% strain compared with the
13 initial case, demonstrating excellent strain-insensitive and potential applications as
14 flexible electronic circuits. The durability and stability of GP₃₀₀@filament is
15 measured under the periodic loading/releasing cycles of 10% strain with 4000 cycles
16 in Figure 5d, the GP₃₀₀@filament could still possess good stability after 4000 cycles
17 with only 0.4% increasement of electrical resistance compared with the initial value.
18 The SEM images of PU filament before and after 4000 cycles are compared and
19 exhibit merely changes of the worm-shape graphene microlayer indicating excellent
20 durability of our PU filament. Furthermore, the abrasion property of GP₃₀₀@filament
21 is also evaluated Figure S4. After 1000 times robust abrasion, the worm-shape
22 graphene micro-layer can still exist indicating good abrasion ability, but after 5000

times robust abrasion, the microlayer was destroyed. According to the basic wearing demand of textile, such abrasion performance is good enough for common wearing textiles. In addition, plucking is also a common action for human motion, so the electroresponse of GP₃₀₀@filament under plucking deformation is shown in Figure 5e. The filament was stretched under 141.4% strain with 45° angle, and then was suddenly released to the free state with attenuation vibration, the resultant relative resistance changes is as low as 0.05 implying the low sensitivity to the external stimulation. In a word, GP₃₀₀@filament reflect remarkable durable and stable strain-insensitive behaviors, which can be applied for the potential wearable electronic circuits.

Figure 5. Strain-insensitive performance of such stretchy electronics. (a) Stress-strain curves of the GP₃₀₀@filament for different strains, showing hysteresis response during the stretching and releasing. (b) Relative resistance changes of GP₃₀₀@filament as a function of stretching (blue line) and releasing (red line) within 400% strain. The insets show the relative resistance

change at the range of 0%-100% and 160%-250% respectively. (c) I-V curves of the GP₃₀₀@filament at different stretching strain levels (from 0 to 300%). (d) Durability and stability test of GP₃₀₀@filament in the process of 10% strain during 4000 loading/releasing cycles, insets: the magnified diagram of the selected cycles, and SEM images of the GP₃₀₀@filament before and after durability test. (e) Relative resistance changes of GP₃₀₀@filament in the pluck of 141.4% strain.

To demonstrate the potential application of strain-insensitive GP₃₀₀@filament as stretchy electronics. As the proof-of-concept, we design a series of preliminary trails. First, GP₀@filament and GP₃₀₀@filament are selected as the elastic electrical circuits for the weighing system at the same time in Figure 6a, 200 g weights is put on the pressure sensor, and two elastic circuits are separately stretched under 10% and 20 % strain, and the $\Delta R/R_0$ curves of each circuits stretching are illustrated in Figure 6b and the $\Delta R/R_0$ change of GP₃₀₀@filament is negligible while that of GP₀@filament reflects regularly changes under the periodic strain. The second trails are designed for an intuitive LED system to observe the luminance change of the LED when GP₀@filament and GP₃₀₀@filament are stretched separately (Figure 6c, d and Supplementary Movie 1). The luminance of LED is barely changed with 300% strain of GP₃₀₀@filament, because $\Delta R/R_0$ is only 0.29 at 300% strain (Figure 6d). In contrast, when the GP₀@filament is slightly stretched, the LED luminance immediately tend to darken at 16.7% strain and extinguished at 33% strain, due to the rapid increasing of $\Delta R/R_0$ (Figure 6d).

Furthermore, the strain-insensitive GP₃₀₀@filament is sewed into fabric substrate to simulate various robust deformations such as stretch, knead, twist, and roll during wearing or using (Figure 6e and Supplementary Movie 2). Besides, the fabrics equipped with GP₃₀₀@filament could also be wore on human wrist, under the wrist joint movement with various bending angles (Figure 6f, g and Supplementary Movie 3), the resultant relative resistance changes is as low as 0.05 result in the luminance of LED still remains brightness and is nearly no disturbed by the elongation of flexible electronic circuits, with the aid of the remarkable the strain-insensitive of GP₃₀₀@filament. Therefore, the superelastic conductive polyurethane filaments with wore-shaped graphene microlayer can be acted as the recommendable candidate as wearable electrical circuit for signal transmission.

Figure 6. The potential applications of strain-insensitive stretchy electronics. (a) - (b) Application of a simple scales using the $GP_0@$ filament and $GP_{300}@$ filament as the stretchable electronic circuits. (c) - (d) Photographs of the LED using the $GP_0@$ filament and $GP_{300}@$ filament as electronic circuits. (e) - (g) The photographs and relative resistance changes of the $GP_{300}@$ filament weave into fabric to conduct signals with intricate motions (e) and under robust wrist joint movements (f, g).

CONCLUSION

Bioinspired from worm micro-structure, we designed and fabricated a special worm-shape GP microlayer $GP_x@$ filament by in-situ pre-stretching wet chemistry deposition. The unique GP microlayer structure endowed the $GP_{300}@$ filament with large reversibility electrical response range of 815% strain, in which the strain-insensitive range was up to 300% with a high Q of 11.26. The high Q provides the possibility for various potential applications in deliver and transmit signals. Additionally, the excellent durability over 4,000 loading/releasing cycles can serve as strain-insensitive stretchy electronic in the long-term used fabric. A series of simple appliances such as scales, LED systems and weaved into fabric to conduct signals under robust wrist joint movements, reveal the feasibility as strain-insensitive stretchy electronics. The proposed conductive filaments combine low-cost fabrication and outstanding performances, providing a direct pathway for strain-insensitive stretchy electronics.

METHODS

Materials. Large size scale flake graphite (lateral size 50 μm) was purchased

1 from Qingdao Tianyuan graphite Co. LTD. Polyurethane (PU) Multifilament
2 (2500D/127f) was purchased from Zhejiang Wally Spandex Co., Ltd. N-Methyl
3 pyrrolidone (NMP) and Polyvinyl Pyrrolidone (PVP) were purchased from Aldrich.
4 Polyurethane solution (PU) with 30 wt.% was purchased from Guangzhou Guanzhi
5 New Technology. Co., Ltd. The water used in this experiment was deionized.

6 **Large scale few-layer graphene inks by pressure homogenizing method.** The
7 few-layer graphene dispersion was exfoliated via the high-speed laminar flow
8 generated by a facile pressure homogenizing route according to the previous works.^{32,}
9 ³³ Figure S1 illustrates the homogenizing mechanism of the large size scale flake
10 graphite into few-layer graphene by a lab-scale pressure homogenizer (AH-2010,
11 ATS Engineering Inc., USA). In details, 0.5 g graphite was added to 100 mL mixture
12 solution of NMP/water (volume ratio 4:1), and 0.01 g polyvinyl pyrrolidone (PVP)
13 served as the surfactant was added into the solution subsequently, following further
14 vigorous ultrasonic treatment 600 W for 30 min until achieving uniform graphite
15 dispersion. The obtained graphite dispersion was gradually injected into the
16 homogenizer with low speed 20 mL/min, and exfoliated at a fixed pressure 50 Mpa,
17 the collected dispersion was cooled in 20°C for 10 min, and the homogenizing process
18 was carried out eight times. After homogenization, the dispersion was centrifuged at
19 5000 rpm for 10 min to separate the thick graphite. And the top two thirds of the
20 dispersion were extracted and retained for use. Such method is a facile physical
21 procedure with low cost, environment-friendly without any chemical treatment.

22 The evolution process from graphite to graphene in the homogenizer (axial

1 symmetry) is shown in Figure S1b, one homogenizing cycle is composed of two
2 motions, loading and releasing pressure. Large size scale flake graphites enter a close
3 container with a high pressure and are entirely squeezed, once the valve moves to
4 leave a current-limiting gap, the pressure is instantaneously released and drove
5 graphites to dash on the impact ring. Such high gradient pressure loss and drastic
6 collision generate the coupled effect of void effect, knock-on effect, and shear effect
7 resulting in the effective exfoliation of graphite. After a series of homogenizing
8 cycles, the interlaminar force of the exfoliated graphite is gradually weakened, and
9 finally evolves into few-layer graphene nanosheets.

10 **Fabrication of conductive polyurethane filaments with worm-shape**
11 **graphene microlayer.** Graphene aqueous solution was first replaced by DMF with
12 centrifuging the dispersion in water at 8000 rpm for 10 min to collect the sediment.
13 The obtained graphene solid was dispersed in DMF with the concentration (10
14 mg/mL). And then, the graphene solution was dropwise added into PU solution under
15 vigorous stirring for 2 h until the mixed solution became homogeneous. We chose
16 30% as the optical case with good film-forming properties and reasonable electrical
17 conductivity, more details can be found in Figure S2.

18 We in-situ deposited the laminate of a GP nanocomposite sheet conductor onto
19 the stretched PU filaments with a worm-shape graphene microlayer on a PU filament
20 core. The process for preparing a GP_x @filament is illustrated in Figure 1a. First, two
21 opposite ends of a PU multifilament were attached on a frame and then the PU
22 filament core was highly stretched under the fabrication strain $x\%$. Afterwards, the

1 stretched PU filaments was gradually immersed into the graphene/PU inks for 5 min
2 until entirely absorption, the soaked PU filaments were moved into the coagulation
3 water bath, the graphene/PU sheet could be in-situ deposited on the surface of the PU
4 filaments with the effect of non-solvent-induced phase separation between DMF and
5 water. After completion of solidification, the strain in the stretched PU filament was
6 gradually released to restore the non-stretched GP_x@filament. In our work, the PU
7 multifilament under seven pre-stretching levels (x equals to 0-300 with 50 interval)
8 were fabricated and characterized. Based on our pre-strain preliminary experiments,
9 300% case was chosen as the pre-strain limit. Once the pre-strain beyond 300%
10 (350% as example in Figure S3), the microlayer of 350% case is partially burst which
11 was caused by the robust resilience force from the axial expansion of internal
12 releasing PU filaments.

13 **Characterization.** The morphological structure of the graphene exfoliated by
14 pressure homogenizing, PU multifilaments, and GP_x@filament samples were
15 characterized via a transmission electronic microscopy (TEM, JEOLH-7650, Japan)
16 and scanning electron microscopy (SEM, EHT: 3.0 kV, work distance: 8.0 mm,
17 ZEISSEVO18, Germany), respectively. Fourier transform infrared spectra (FTIR) of
18 the samples were measured on a Nicolet 5700 FTIR spectrometer (Thermo Nicolet
19 Corp, USA), the wave number range was 4000-500 cm⁻¹. GP₃₀₀@filament was rubbed
20 5000 cycles by LFY-109A Fiber Abrasion Tester, the particle sizes of the abrasive
21 paper in abrasion test is P1000 and the tension is 100N which can simulate robust
22 abrasion of garment.

All the sensing characterizations of the GP_x@filament were evaluated using the System Source Meter (Models 2601B, Keithley, USA). The dynamic strain-stress behaviors of the filaments were measured with a tensile testing machine (Instron 4206, Instron Ltd., UK) at the initial test length of 2 cm and at a stretch rate of 6 mm/min.

The quality factor (Q) of the GP_x@filament can be calculated as follows,

$$Q = \frac{\Delta L / l_0}{\Delta R / R_0} \quad (1)$$

where R_0 represents the original electrical resistance, ΔR and ΔL indicate the change of resistance and length after the corresponding strain.

ASSOCIATED CONTENT

Supporting Information

Figure S1: Lab-scale pressure homogenizing method preparing few-layer graphene;

Figure S2: The optical case of graphene/PU nanocomposite (GP) inks with various graphene weight ratios;

Figure S3: SEM images of GP₀@filament, GP₃₀₀@filament, and GP₃₅₀@filament;

Figure S4: The abrasion property of GP₃₀₀@filament;

Figure S5: Models for the GP microlayer of GP_x@filament with various pre-stretching;

Supplementary Method 1: Theoretical analysis of the filament resistance after pre-stretch;

Supplementary Method 2: Theoretical analysis of the filament resistance on strain.

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