Stretchable Conductive Fibres of Ultrahigh Tensile Strain and Stable Conductance Enabled by a Worm-Shaped Graphene Microlayer

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Abstract. To enable stable conductance for smart wearable devices, we designed a worm-shaped stretchable conductive fibre by coating graphene/polyurethane microlayer onto prestretched polyurethane (PU) filaments. Such fibre can be stretched up to 1010% with a wide reversible electroresponse range ($0 < \varepsilon < 815\%$), long-term durability (>4000 stretching/releasing cycles), good initial conductivity ($\sigma 0 = 124$ S m–1), and high quality factor (Q = 11.26). Remarkably, it shows distinctive strain-insensitive behaviour ($\Delta R/R0 < 0.1$) up to 220% strain, thereby can provide a direct pathway for stretchy electronics.

Background

A smart wearable device, as an important case of artificial intelligence, is a complex microsystem composed of electronic skin [1], actuators [2], stretchable electronic circuits, flexible supercapacitors [3], strain sensors [4] and so on. The conductivity of most reported stretchable conductive fibres will change if stretched or strained, suitable as sensors. But to ensure the stable performance and provide satisfying wearing experience, strain-insensitive electronics with good stretchability and durability are also essential for wearable devices. In present work, we designed a worm-shaped strain-insensitive conductive fibre by coating graphene/polyurethane microlayer onto prestretched polyurethane filaments. Here, we discuss its fabrication, testing results and geometry.

Results

The worm-shaped graphene coated polyurethane (PU) was prepared as shown in Fig. 1. Briefly, the pristine PU filament was stretched under the fabrication strain x% and then immersed into the graphene/PU inks for capillary absorption; afterward, the graphene/PU microlayer could be in situ formed on the soaked PU filaments in the coagulation water bath. After releasing the prestretching forces loaded on the filament substrate, the worm-shaped graphene microlayer was achieved.

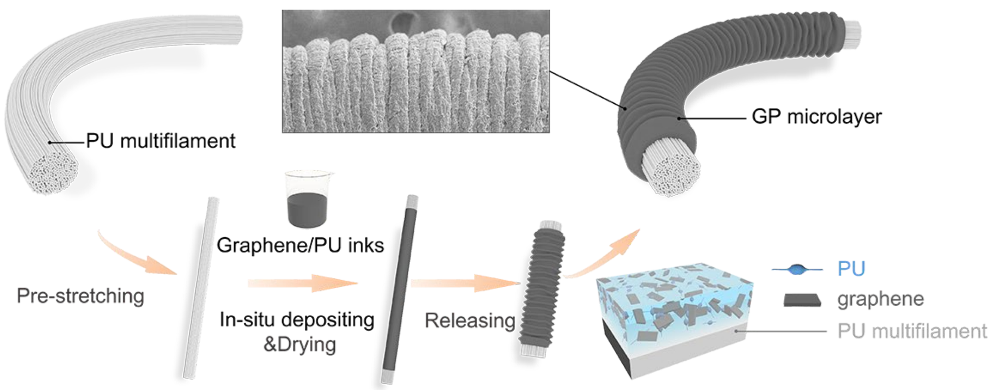


Fig. 1 Schematic diagram of preparing the worm-shaped filaments.

The strain-insensitive performance of graphene coated polyurethane fibre with 300% pre-stretching rate (GP300@filament) was investigated in Fig. 2. As shown in Fig. 2a, its relative resistance changes remain under 0.8 under the wide strain range 0–400% with hysteresis as small as 0.02, indicating outstanding strain-insensitivity and repeatability. The durability and stability of GP300@filament is measured under the periodic loading/releasing cycles of 10% strain in Fig. 2b, which could still possess good stability with only a 0.4% increase of electrical resistance after 4000 cycles. In addition, the electroresponse of GP300@filament under plucking deformation is also investigated in Fig. 2c. The resultant relative resistance change is as low as 0.05, implying the low sensitivity to the external stimulation. In a word, GP300@filament reflects remarkable durable and stable strain-insensitive behaviours, which can be applied for the potential wearable electronic circuits.

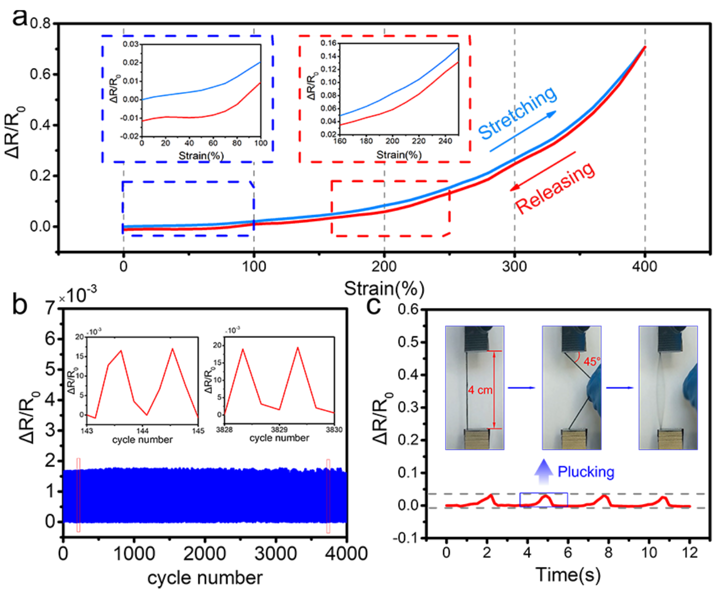
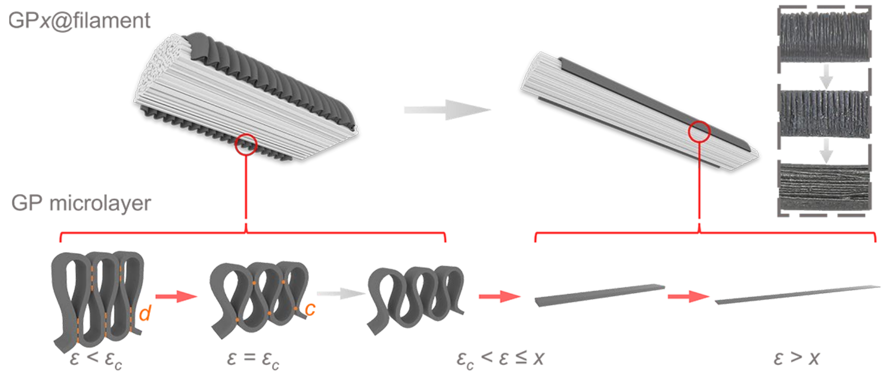


Fig. 2 Strain-insensitive performance of such stretchy electronics. (a) Relative resistance changes of GP300@filament as a function of stretching (blue line) and releasing (red line) within 400% strain. (b) Durability and stability test of GP300@filament in the process of 10% strain during 4000 loading/releasing cycles. Insets: the magnified diagram of the selected cycles and SEM images of the GP300@filament before and after durability test. c) Relative resistance changes of GP300@filament in the pluck of 141.4% strain.

In order to further clarify the mechanism of electrical conductive of worm-shaped graphene/PU microlayer, the micro geometrical model of the GP microlayer unit on GPx@filament during the stretching process is illustrated in Fig. 3. The stretching process is classified into three simple stages, which can be divided by two boundary points. The contact critical point ($\varepsilon = \varepsilon c$) is the special boundary where the two legs of the GP layer loop are in point-contact, and the other boundary is the straighten prestretching state ($\varepsilon = x$) that the strain degree matches with the prestretching state. Therefore, the tendency of the microstructure of GP microlayer generally passes through three phases: the original state to the contact critical point ($0 < \varepsilon \le \varepsilon c$), the contact critical point to the straighten prestretching state ($\varepsilon c < \varepsilon \le x$), and the state after prestretching ($\varepsilon > x$). The total resistance during different stages is expressed by equations shown in Fig. 3. The measured electrical conductivity of GP300@filament could almost match such model but is slightly smaller than the calculated result. This could be due to its less ordered structure of PU filaments and less regular worm-shaped graphene/PU microlayer compared to the theoretical simulation. Therefore, the discrepancy could occur between theoretical and experimental results, but within a reasonable margin of error.



$$R = \frac{2R_l l_0}{2l_0 + \pi[2A - (x\% - \varepsilon\%)l]A\sigma_d D_0 R_l} \qquad R = R_l \qquad R = \frac{(\varepsilon\% + 1)^2}{(x\% + 1)^2} R_l$$

Fig. 3 Geometrical model of the GP microlayer unit on GPx@filament and the corresponding theoretical formula at different stages during the stretching process.

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