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Auxiliary soft beam for the amplification of the elasto-capillary coiling: Towards stretchable electronics

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ABSTRACT

A flexible fiber carrying a liquid drop may coil inside the drop thereby creating a drop-on-fiber system with an ultra-extensible behavior. During compression, the excess fiber is spooled inside the droplet and capillary forces keep the system taut. During subsequent elongation, the fiber is gradually released and if a large number of spools is uncoiled a high stretchability is achieved. This mechanical behaviour is of interest for stretchable connectors but information, may it be electronic or photonic, usually travels through stiff functional materials. These high Young's moduli, leading to large bending rigidity, prevent in-drop coiling. Here we overcome this limitation by attaching a beam of soft elastomer to the functional fiber, thereby creating a composite system which exhibits in-drop coiling and carries information while being ultra-extensible. We present a simple model to explain the underlying mechanics of the addition of the soft beam and we show how it favors in-drop coiling. We illustrate the method with a two-centimeter long micronic PEDOT:PSS conductive fiber joined to a PVS soft beam, showing that the system conveys electricity throughout a 1900% elongation.

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1. Introduction

At small scales capillary forces may be used to fold elastic structures, and several studies have shown how one can design shape and form in microstructures by the careful utilization of surface tension, provided Young's moduli are low enough or structures are thin enough [1–5]. The mechanics and physics of drops on fibers have been studied from the point of view of wettability and mist capture [6–9] but recently, finding inspiration in spider webs [10–12], we have seen the introduction of elastocapillary drop-on-fiber systems where the flexible fiber coils inside a liquid drop. This spooling mechanism provides the system with a constant force behavior under extension (*i.e.* vanishing stretching modulus) in addition to extreme extensibility [2,13-15]. In-drop coiling basically requires thin fibers and up to now ~10μm diameters and ~10 MPa materials have been successfully used. With the aim to design functional microstructures utilizing in-drop coiling, we turn to the use of metalic materials with the drawback that functional materials usually have high Young's moduli [16] (metals for electronics, glass for photonics, for which

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$E \sim 50 - 100$ GPa). For these materials, if the bending rigidity is to be kept low, the structure has to be dramatically thin, in the order of nanometers. Yet thin structures are difficult to manufacture [17,18], and lose their functionality (electric resistance of a fiber decreases quadratically with decreasing radius).

Interest in the last decade in flexible electronics is driven by applications in consumer electronics (*e.g.* flexible laptop or smartphone screens), but also integration in the human body to design sensors that can be implanted on living organs [19]. If bending flexibility is achieved by designing thin enough structures, stretching compliance is usually obtained through the use of wavy structures; in both cases the relative rigidity of the material (silica) is dealt with by structural design. Some examples where the focus has been on the material side are the use of conductive polymers in the fabrication of conductive elastic materials [20]. Here we use in-drop coiling to design highly extensible fibers. Moreover we introduce the use of an auxiliary soft beam that helps in-drop coiling of these elastic fibers, and show that coiling can be achieved with electrically conductive materials.

The paper is organized as follows. In Section 2 we give thorough information on experimental methods and materials used. In Section 3 we recall the conditions under which an elastic fiber may coil inside a liquid drop. We then point out in Section 4 that gravity inevitably hinders the coiling procedure, but in Section 5 we introduce the use of an auxiliary soft beam which on the contrary enhances this coiling procedure and enables us to show in Section 6 the in-drop coiling of a conductive relatively stiff fiber. We discuss our results in Section 7 and conclude.

2. Materials and methods

Different materials were used to fabricate the fibers in order to cover a large span of Young's moduli in the experiments (see Table 1). The thermoplastic polyether polyurethanes (TPU) fibers were fabricated from pellets (BASF) following a manual melt-spinning process described in [21] where a TPU pellet is molten at 225 ° C on a hot plate and then touched by the tip of needle. Pulling the needle rapidly then draws out a microfiber which immediately solidifies in the air, leaving the operator with a meter long microfiber. The different TPU's Young's moduli were measured through a force displacement test using the Femto-Tools micro-forces instrument FT-MTA02 (capacitive deflection measurement).

Polylactic acid (PLA) is commonly used for 3D printing, its Young's modulus was measured using a Shimadzu AG-X Plus electromechanical test frame on a PLA wire of radius 1.5 mm. Just as for the TPU, PLA fibers were fabricated with a melt-spinning technique: a small amount of PLA was molten on a hot plate (290 ° C) and subsequently pulled out rapidly with the tip of a needle, thus creating the solid microfiber.

Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, a conductive polymer) dry re-dispersible pellets from Sigma-Aldrich were used to draw fibers using the wetspinning process described in [22,23]. A 2.9 wt.% PEDOT:PSS-water solution was first prepared using a Mettler-Toledo MS scale. The solution was then injected through a calibrated needle (Adhesive Dispensing Ltd and Cluzeau Info Labo) out of a Terumo 10 mL glass syringe into an acetone bath. As water is more soluble in acetone than in PEDOT:PSS, it flows out of the PEDOT:PSS, which itself is not soluble in acetone, and the injection leads to the contraction and solidification of a continuous flow of PEDOT:PSS. The resulting solid microfiber was then manually extracted from the acetone bath and dried out in ambient air for about one minute. Even though the PEDOT:PSS fibers thereby created are not exactly the same as in [23], we use their value for the Young's modulus. Our PEDOT:PSS fibers showed a specific electric conductivity of 7 S/cm.

Table 1
The materials used for fiber fabrication and their respective Young's modulus E.

Material	Young's modulus E
PVS shore 8	200 ± 30 kPa
TPU E1170A	9.50 ± 2 MPa
TPU E1185A	23.6 ± 4 MPa
TPU E1198A	66 ± 10 MPa
TPU E1174D	560 ± 40 MPa
PEDOT:PSS	2.5 ± 0.7 GPa
PLA	2.7 ± 0.7 GPa

End-to-end electrical resistance of straight and in-drop coiled PEDOT:PSS microfibers were measured using an ohmmeter (Amprobe 5XP-A). The electrical connections between macro-parts and the microfiber were established with a Hi-Bond Conductive Copper Tape.

No active control on the fiber's cross-section radius is possible with both of the aforementioned fabrication techniques, but it was optically measured after its fabrication for each fiber using a Leica macroscope (VZ85RC) with a Leica DFC-295 camera. The optical resolution of this macroscope is of the order of magnitude of the diameter of the thinnest fibers that were used (around 2 μm). Since the bending rigidity of a cylindrical fiber is proportional to the fourth power of the radius of its cross section, the fibers' radii are a key parameter in the experiments. Therefore an algorithm was written to take into account the blurriness of the image near the fiber edges, and after image post-processing the absolute uncertainty on the fiber's radii was lowered to 0.5 μm.

Zhermack Shore 8 Polyvinyl-siloxane (PVS), was used to fabricate the soft auxiliary beams. The following technique was used to obtain ~ 2 cm long fibers with ~ 100 μm thin cross sections. Before reticulation, the PVS was poured on the 170 μm wide side of a microscope glass slide (VWR, 22×50 mm, thickness no 1.5). A small part of the PVS then sat on the side when reticulation starts, whereas the surplus ran off the edges. After reticulation ($\approx$ 20 min at ambient temperature), a fiber is extracted from the side of the glass slide. Its cross section resembles a circular cap which we will here, for simplification, consider to be rectangular of height and width equal to those observed on the circular cap. These geometric dimensions were observed optically with the Leica macroscope. The aspect ratio $k = \frac{h_{\text{soft}}}{w_{\text{soft}}}$ of the cross-section of the fiber was found to be $k = 0.34$. The perimeter and the quadratic moment of inertia of the cross section were also approximated to be those of a rectangular cross section. Finally, the Young's modulus of our PVS was determined by a force-displacement measurement on a cylindrical fiber of radius 750 μm and length 8.5 cm.

The droplets used for the experiments are silicone oil droplets (viscosity 100 cSt at 25 ° C) from Sigma-Aldrich. It is to be mentioned that this silicone oil does not swell TPU, PLA and PEDOT:PSS, but does swell PVS. As a first order approximation, no Young's modulus rectifications were applied to the PVS beams. However, characteristic dimensions of these PVS beams increased by about 20% due to this swelling. Silicone oil was found to perfectly wet all of the used materials (Young-Dupré equilibrium wetting angle $\theta_Y = 0$) which is why we consider $\gamma_{SV} - \gamma_{SL} = \gamma\cos\theta_Y = \gamma$, where γ_{SV}, γ_{SL} and γ are respectively the Solid-Vapor, Solid-Liquid and Liquid-Vapor specific interface energies.

In order to determine whether a fiber (with or without auxiliary soft fiber) was 'coilable' in a silicone oil droplet, about 10 droplets of different sizes were put on the fiber, seeking the best droplet size (big enough to allow coiling, but not too big for it not to be too heavy). If the fiber undergoes at least 2 coils inside one of the drops upon compression of its ends, at whichever hanging angle, the fiber is considered 'coilable'. If no droplet leads to this type coiling, the fiber is considered 'uncoilable'. Following this procedure, a fiber would first be tested alone (without auxiliary soft beam), and then would be affixed to the auxiliary soft beam and the composite fiber would be

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