



Energy harvesting and movement tracking by polypyrrole functionalized textile for wearable IoT applications

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ABSTRACT

Textiles for health and sporting activity monitoring are on the rise with the advent of smart portable wearables. The intention of this work is to design wireless monitoring wearables, based on widely available textiles and low environmental impact production technologies. Herein we have developed a polymeric ink which is able to functionalize different types of textile fibers (including silver conducting fibers, cotton, and commercial textile) with polypyrrole. These fibers were weaved together with a thinner silver conducting fiber and carbon fiber to form a touch-sensitive energy harvesting system that would generate an electric output when mechanical pressure is applied to it. Different prototypes were manufactured with loom weaving accessories to simulate real textile cloths. By simple touch, the prototypes produced a maximum voltage of 244 V and a maximum power density of 2.29 W m⁻². The current generated is then transformed into a digital signal, which is further utilized for human motion or gesture monitorization. The system comprises a wireless block for the Internet of Things (IoT) applicability that will be eventually extended to future remote health and sports monitoring systems.

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1. Introduction

The rapid expansion of Internet of Things (IoT) technologies has unlocked new opportunities across various industries, with wearable devices standing out as a dynamic frontier in this evolving landscape. Wearables are gaining significant traction due to their diverse applications across sectors such as healthcare, fashion, smart packaging, and military operations. Their ability to integrate seamlessly into daily life while delivering real-time data and connectivity underscores their transformative potential. As the IoT ecosystem advances, wearables are poised to drive innovation, offering cutting-edge solutions that enhance user experience and operational capabilities across multiple consumer-focused domains.

1.1. Sustainable “on-use” energy

An endless energy source that needs to be portable and flexible is required. The battery is still the primary option for portable energy sources, but it requires recharging, periodical maintenance, or replacement [1–3]. Effectively, self-powered technology has a great impact due to its ability to work independently without any other external energy suppliers. Such self-powered devices [4–7] are integrated with energy harvesting and transducing systems, where solar [8,9], humidity [10,11], thermal [12–14], fluid flow [15,16], or mechanical energy [17–20] is converted into electrical energy. Mechano-responsive energy harvesters can be used spontaneously without using a storage device, which can be very useful for wireless signal mechanisms or tracking systems [21,22]. Concurrently, mechanical energy harvesting has gathered interest in the scientific community because of its diverse sources and huge availability in our daily lives (such as vibration, pressure or motion, or any human interaction) [17,23–26], giving use to a lot of dissipated energy that would otherwise be wasted. Piezo and

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triboelectric materials have been the most explored materials and are still the big tendency in mechanical energy harvesting investigation [27,28]. When pressure is imposed onto the surface of such materials, an electrical output is generated due to contact electrification and electrostatic induction [29]. Therefore, a new approach should be conceptualized whether it is technical or materialistic evolution, to adapt them as powerhouses of “on-use” energy for future IoT-based wearables.

1.2. E-textile conformability

Although the idea of e-textiles has been around for decades, it has only been emerging in the last few years [30–35]. Mostly, e-textiles are progressing through the integration of miniaturized electronic components into textiles [36,37]. However full integration of electronics into textiles is itself challenging, due to the difficulty in ensuring device efficiency, often involving the usage of bulky electronics and wired connections, while keeping textile properties such as conformability and durability. Another developing vector can be to exploit the textile itself, associated with nanotechnology through functionalization and designing the products in form of woven, knitted, or non-woven structures of traditional fibers; so that, the textile can act as an integral part of device engineering and take part in direct interaction [38,39]. Due to the specific features of textiles such as flexibility, lightweight, and wearability, as well as their intrinsic warmth and fit properties, electronic textiles seem to be highly appropriate to be implemented in the next generation of smart electronic communications and energy harvesting applications.

1.3. Polypyrrole as active material for energy harvesting textiles

Given that, the functionalization of textiles with conjugated polymers (CPs) can be a key enabler owing to the current development of these polymers in energy harvesting [40–42]. CPs are organic macromolecules with delocalized π -electrons arising from the overlapping of p -orbitals. These delocalized π -electrons result in good charge transference between the polymeric chains, which explains the high conductivity of the polymer. In addition, conjugated polymers are an attractive option for new green electronic devices because of their flexibility, ease to control electrical and chemical properties through doping-dedoping strategies, low-cost, and elementary synthesis [43]. In the field of smart materials, CPs, have brought about a lot of interest, in comparison to other polymers, due to their multi-functionality. They have been applied as active materials in pressure sensing devices, gas sensing, energy harvesting, dielectric in supercapacitors, displays [44–50], as well as electrode materials in several types of smart and flexible electronics and in the field of displays (with the advent of polymer light-emitting diodes), supercapacitors, transistors, sensors, data storage devices [51–58], etc. Moreover, conjugated polymers also exhibit promising biocompatibility, which opens possibilities for its use in biomedical applications, such as wearable health monitoring sensors that come into direct contact with human skin [59]. Its non-toxic nature ensures that it can be safely incorporated into wearable devices without adverse effects on the user [60].

The idea behind the present work is to use the mechanical energy harvesting abilities of polypyrrole (PPy) for sustainable IoT exercise monitoring applications in wearables. Therefore, we have functionalized the commercially available silver-coated conductive textile fibers (CTFs), cotton fiber (CF), and commercial glove textile (CGT) with PPy by a very simple chemical route. Additionally, PPy has good adhesion with organic and metallic materials. The inks manufactured herein are water-based and able to functionalize different types of textiles, including cotton, metallic fibers, polyester, etc. Its adaptability to several types of substrates makes them a good can-

didate for their adaptation to industrial products, in a sustainable way. The functionalized fibers were then fabricated into an energy harvesting device that was applied for as a wireless monitoring (body movements) system through a properly developed interface/-communication IoT system, with capability of high data gathering at $115200 \text{ bits s}^{-1}$ as well as filtering and data interpolation.

2. Results and discussion

2.1. Synthesis and chemical analysis

PPy was synthesized by oxidative chemical synthesis (OCS). Reasons behind the choice of the OCS approach include the easy tuning process of its electrical and morphological properties and elementary synthesis that does not require any cleanroom or other complex techniques. Two precursor solutions were prepared for the functionalization of textile fibers with PPy. (i) An oxidant solution contained ammonium persulfate (APS) and HCl (37%). HCl was chosen as primary dopant, for its wide report in increasing the conductivity of PPy by several orders of amplitude. The presence of HCl in the reaction will provide a Cl^- which will increase the ionic strength of the medium, resulting in a tendency for agglomeration of PPy's granular structured particles. In addition, it will induce the formation of polycations, influencing the creation of a polaronic conduction band. HCl is a good alternative for increasing the conductivity of the polymer without increasing too much the concentration of oxidizing agent, which tends to create less stable structures [61]. (ii) A monomer solution contained pyrrole monomer in deionized (DI) water. The fibers were then strapped to the inner wall of the reaction vessel with Kapton tape and the whole system was placed inside an ice bath. Both solutions were consecutively dropped into the beaker containing the fibers and the reaction was let to occur uninterruptedly for different time durations (30, 60, and 90 min). The functionalization time proposed is related to the characteristics of the synthesis and the nature of the polymeric reaction. The polymerization has origin in the combination of several reduced and oxidized blocks of aniline monomer. It is possible to regulate the amount of oxidized and reduced monomers in the polymer through controlled synthesis parameters, which determines the different phases of polypyrrole, depending on their oxidizing state. For our purposes, it is important to have polypyrrole close to the more balanced oxidized and reduced state (the conductivity can be tuned with doping dedoping processes) due to its semi-conducting properties. For our synthesis parameters, the formation of this phase occurs in functionalization times between 20 and 100 min of reaction. By changing the synthesis parameters, it would be possible to decrease the functionalization time, for instance, by realizing the synthesis at room temperature, or by accentuating the concentration of the oxidizing agent. This would result in shorter periods of induction resulting in chaotic agglomeration of PPy nucleates and shorter polymer chains, consequently leading to less conductive and less stable polymers [62,63]. The purpose was to study the influence of polymerization time on the electrical and morphological properties of PPy. Once the synthesis was completed, the PPy-functionalized textile fibers (FTFs) were removed from the vessel and washed to remove any excess material and left to dry at ambient temperature. Fig. 1(a) presents a simple schematic of the above-mentioned synthesis procedure. A uniform black color (which is basically a typical characteristic of PPy) was observed around the fibers, being an indication that the polymerization process was completed. Only the upper end of the fiber that was tapped with Kapton tape had not been functionalized (this was intended to access later the inner Ag core of the functionalized fiber as an active electrode of the device). After taking out the

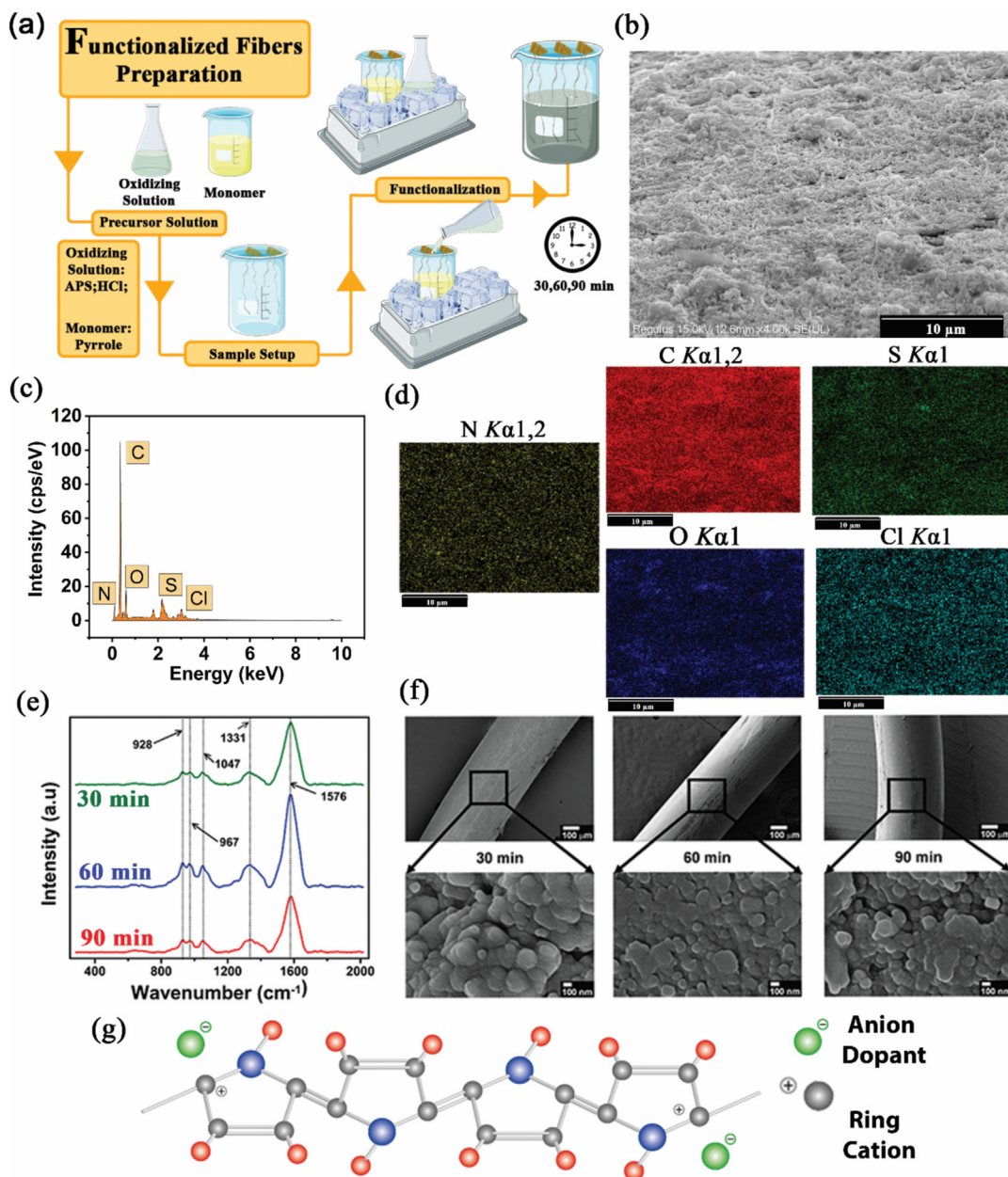


Fig. 1. (a) Schematic of the functionalization of silver fibers with PPy. The oxidant solution and the monomer solution were brought to ice bath temperature and then added to a vessel containing the fibers and the synthesis time proceeded at low temperatures for 30, 60 and 90 min; typical EDS of the functionalized fibers (sample: FTF prepared with 60 min functionalization) of (b) EDS layered image, (c) EDS spectrum elemental mapping, and (d) EDS elemental mapping; (e) Raman spectra of the functionalized fibers for different synthesis durations; (f) FESEM images of the PPy structures obtained for each synthesis time; (g) the bi-polaronic structure of PPy, where the double cations are responsible for its electronic mobility and charge generation mechanism.

fibers from the reaction bath, the remaining solution was subjected to paper filtration and dried in an oven at 60 $^{\circ}\text{C}$ for the next 24 h to collect eventually as-produced PPy powders for further chemical analysis.

The corresponding energy-dispersive X-ray spectroscopy (EDS) spectrum and elemental mapping (using Oxford XMax 150) of the typical PPy functionalized fiber have been shown in Fig. 1(b–d) respectively. Chlorine (^{35}Cl) is also present in great quantity because of the addition of HCl during the synthesis of PPy, which increases the presence of anionic Cl^{-} dopant in the polymeric chain. Carbon (^{12}C), nitrogen (^{14}N), and oxygen (^{16}O) are part of the molecular structure of the Pyrrole monomer, whereas sulfur (^{32}S) is present because the oxidant agent used upon the synthesis of PPy was APS, indicating the presence of the persulfate group.

Fig. 1(e) shows the Raman spectra of the powders collected from different polymerization times. It is clear from the spectra that the chemical phases from all the polymerization times are similar in characteristics and correspond to the successful formation of PPy having all its typical characteristic peaks, as expected. The strongest peak is observed at 1576 cm^{-1} , which corresponds to the symmetric stretching of the aromatic $\text{C}=\text{C}$ ring of PPy, representing the oxidation state. Other peaks are around 928 and 967 cm^{-1} , which are associated with the ring deformation caused by the bi-polarons and the polarons species, respectively. It is possible to observe even more two peaks, one associated with the $\text{C}-\text{H}$ in-plane deformation of the oxidized PPy (1047 cm^{-1}) and the other associated with the ring stretching mode of PPy chain (1331 cm^{-1}) [64–66]. The Raman analysis of the powders

at different functionalization times did not suffer meaningful variations, presenting peaks of similar intensity at the same wavenumber. Field emission scanning electron microscopy (FESEM) was used to visualize the morphology of the PPy nanostructures. Fig. 1(f) depicts the high-resolution image of the conductive textile fibers after functionalization for each individual synthesis time. The image allows us to observe that the conductive fibers were successfully functionalized with a uniform PPy coating, having a cauliflower-like morphology with an average granular size of 80–120 nm. The sizes of the nanostructures observed here are smaller than some others observed in the literature, possibly due to the use of HCl as a dopant, which is one of the smallest dopants that can be used, resulting in more compact structures; whereas the amorphous cauliflower-like morphology is a common granule type for PPy synthesized by OCS [67]. Moreover, no significant morphological difference was noticed for PPy with different polymerization times. The electronic properties of PPy can be associated with the synthesis parameters. Due to its high oxidation potential, APS is added to water, SO_4^{2-} ions are formed, which will then attack the weaker π -electron bonds of the monomer; consequently a free radical and a cation will be formed within the ring, due to the loss of an electron. When higher doping levels are employed, further oxidation will proceed with pairs of polarons merging with each other forming more energetically stable, bi-polarons. The polarons and bi-polarons will act as charge carriers; in the case of PPy, they will form a p-type conductor since there is the creation of a hole due to an electron being lost during the oxidation phase. Fig. 1(g) represents a schematic with a possible representation of the oxidized state. Furthermore, the synthesis was tested for its reproducibility, with several different batches being produced for all the polymerization times. SEM, EDS and Raman analyses were conducted for all the reproduced batches. The results of the reproducibility test are detailed in Figs. S1–S28 in the Supporting Information.

2.2. Architecture and manufacturing design of prototyped devices

The design and fabrication of wearable devices integrated with smart electronics present significant challenges, primarily in balancing functionality with the requirements of the textile industry. The architecture of these devices must not only be appealing and conformable to the needs of wearable technology but must also retain the primary functionalities of the textiles. Herein, various textile-based prototypes have been developed, incorporating both functional and structural elements to meet these demands.

The prototypes are constructed using a two-layer architecture designed to optimize performance while maintaining flexibility and integration with fabrics. The first layer, known as the “active layer”, is fabricated from a conjugated polymer that provides the core functionality of the wearable electronics. Specifically, this active layer is developed using a PPy functionalization process. PPy is a conductive polymer that has been deposited onto different types of fibers, making them suitable for use in wearable applications. The functionalization process is carefully controlled to ensure uniform coating and optimal conductivity, while still maintaining the mechanical properties of the fibers, such as flexibility and durability. The second layer in the architecture is the “charge collector layer”, which plays a crucial role in the operation of the wearable device. This layer serves as an electrode, collecting charge from the active layer during device operation. It is fabricated using a commercial conductive textile, chosen for its high conductivity, flexibility, and ease of integration with other textile layers. The charge collector layer is designed to interface seamlessly with the active layer, ensuring efficient charge transfer without compromising the overall flexibility of the wearable device.

The combination of the PPy-functionalized active layer and the conductive textile charge collector layer results in a wearable

device that is both functional and adaptable to the demands of the textile industry. The integration of these layers into textile-based wearables allows for the creation of smart fabrics that are not only electronically active but also comfortable and practical for everyday use. Through this design, the prototyped wearables successfully merge the worlds of electronics and textiles, paving the way for future advancements in wearable technology. Following this, four different prototype designs have been discussed according to their fabrication processes.

Design 1.

The first prototyped device, referred to as the “energy harvesting fiber” (EHF), was developed using a specialized spiral weaving technique to create a functional and efficient energy harvesting system. This design of EHF involved the use of two different types of fibers: a thicker functionalized textile fiber (FTF) with a diameter of 400 μm and a thinner conductive textile fiber (CTF) with a diameter of 150 μm . The fabrication process began by spirally weaving the thinner CTF around each of the thicker FTFs. The CTF served as the charge collector layer within this double-electrode system, while the uncoated metallic end of the FTF was utilized as the back electrode, forming a well-coordinated and effective energy harvesting structure. This systematic design enabled the EHF to function as a self-contained energy harvesting unit, capable of generating and collecting electrical charge through its fiber-based architecture. The overall architecture of the energy harvesting device involves coupling of several EHF. The arrangement of these fibers in series allowed for the enhanced energy harvesting capabilities, taking full advantage of the surface area and interaction between the fibers. The design was carefully engineered to maintain flexibility, which is critical for wearable applications, without compromising the performance of the device. To ensure the durability and protection of the EHF system, the entire device was encapsulated in a layer of polydimethylsiloxane (PDMS). PDMS is a flexible, biocompatible silicone material that provides mechanical protection while maintaining the conformability of the device. The PDMS encapsulation was achieved by mixing silicone elastomer and curing agent in a mass ratio of 10:1. The curing process was carried out at 60 °C, which ensured a stable and protective coating around the energy harvesting fibers without affecting their flexibility or functionality. The schematic representation and real image of the prototyped EHF device can be seen in Fig. 2(a), showcasing the spiral weaving technique and the overall structure of the system. This design marks an innovative approach to energy harvesting through textile-based electronics, combining advanced materials and fabrication techniques to create a flexible, functional wearable energy solution.

Design 2.

The second prototyped design involves the development of a lab-scale weaving system specifically adapted to produce EHF prototypes. This innovative loom was custom-designed and fabricated using three-dimensional (3D) printing technology, utilizing the Ultimaker 2+ to create the necessary structural components. The purpose of this design was to facilitate the precise weaving of CTFs around FTFs for lab-scale production. In this system, the FTFs were arranged in a snake-like format along the edges of the loom, providing the foundational structure for the prototype. A needle was then used to weave the CTFs around the FTFs in a perpendicular direction, also in a snake-like pattern. This systematic weaving technique allowed for the creation of well-organized, uniform prototypes. Two different types of devices were fabricated using this method.

D-MM (Device with one CTF): This device was composed of a 1-meter-long FTF with a single CTF woven around it. The CTF acted as the charge-collecting electrode, wrapping around the FTF in a consistent and structured pattern, creating a functional energy-harvesting system.

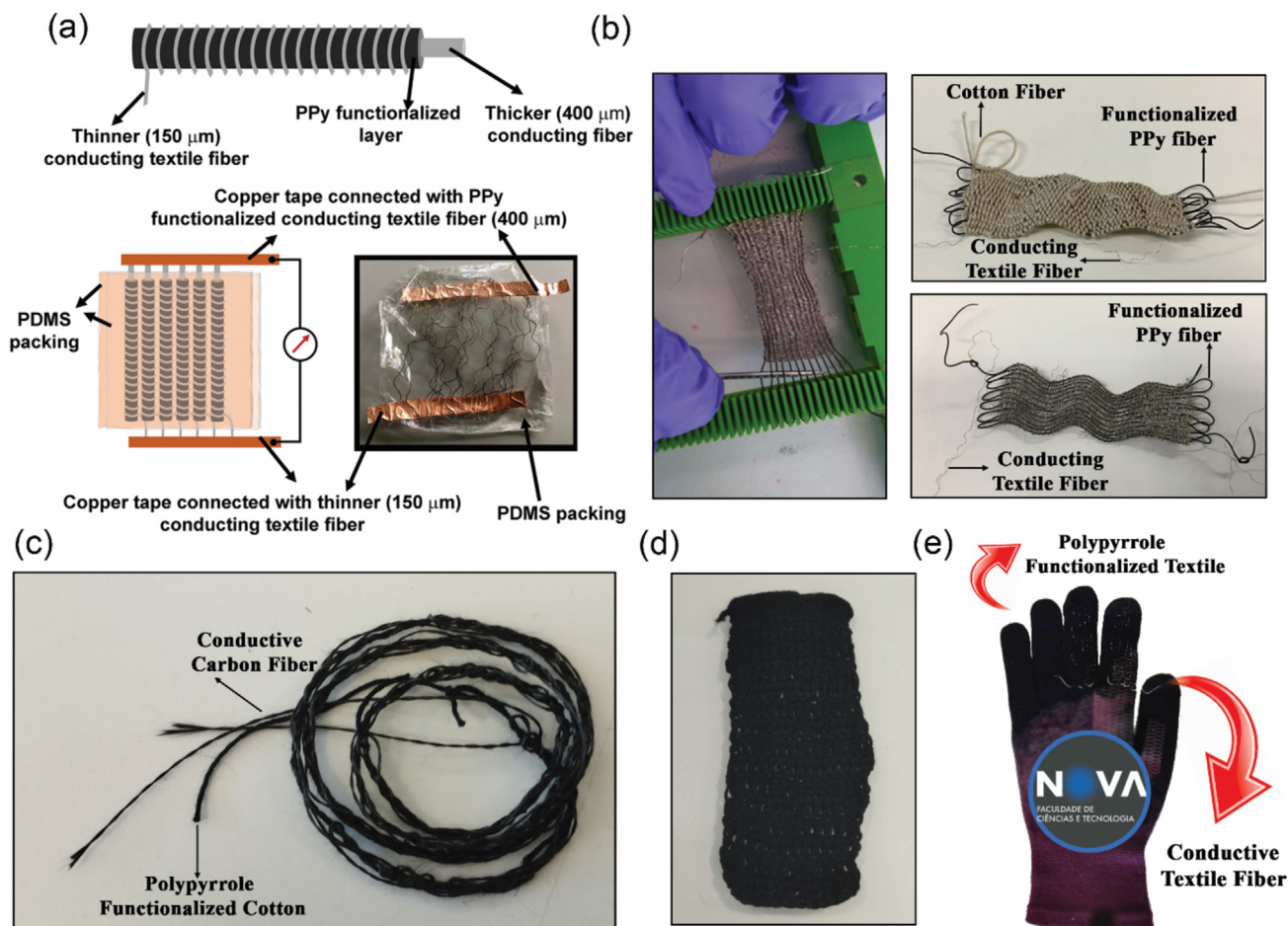


Fig. 2. (a) Schematic of the weaved EHF, making a system that is composed of the FTF and one CTF weaved around it and schematic with snapshot of the devices manufactured with PDMS cover, in this case, the CTF was manually weaved around the FTF. Together the active layer (i.e., FTF) and the charge collector layer (i.e., CTF) are the main contributors to the charge transfer mechanism; (b) digital image of the lab-scale weaving loom used to make the more conventional-looking textile devices and digital images of D-MMC (top) and D-MM (down) device; (c) PPY functionalized cotton and carbon fiber weaved into a single fiber for manufacturing (d) smart textile garment (D-CFC), illustrating the adaptability of crochet techniques for producing wearable electronics; (e) PPY functionalized textile from commercial glove with weaved conductive textile fiber.

D-MMC (Device with CTF and CF): In this device, a similar 1-meter-long FTF was used, but the weaving process alternated between two fibers: a CTF and a cotton fiber (CF). The CTF, serving as the electrode, was woven in a perpendicular direction around the FTF, while the CF was interspersed between the CTFs to simulate real fabric conditions. This design was intended to mimic actual textile properties and test the device performance under realistic usage scenarios.

In both designs, the CTF served as the primary electrode for charge collection, while the CF in the D-MMC design was added to better understand the interaction between functionalized fibers and traditional textile materials. The incorporation of CFs and CTFs into a woven structure offered more flexibility and durability, while the usage of weaving loom facilitated the consistent production of several patterns in a controlled environment. The schematic and real images of this lab-scale weaving loom and the resulting devices can be seen in Fig. 2(b). These designs demonstrate the adaptability of traditional weaving techniques for use in smart textile production, showcasing the potential for scalable manufacturing of energy-harvesting textiles using advanced fabrication methods like 3D printing.

Design 3.

The third prototyped design introduces a new approach, where CFs, traditionally used in the textile, were functionalized to enhance the wearability. In this design, the CFs were chemically

treated to become functionalized cotton fibers (FCF), which acted as the active layer in the wearable device. These FCFs were then carefully wrapped around commercial carbon fiber (CCF), which served as the charge collector layer due to its conductive nature (Fig. 2c). The composite threads created from the combination of FCF and CCF were fabricated into small crochets using traditional crochet techniques and needles, mimicking the structure of wearable fabrics. This crochet technique allowed the fibres to be formed into a flexible, fabric-like material while still maintaining the electroactive properties necessary for the device to function. In this configuration, the FCF served as the active layer, generating electrical charge, while the CCF acted as the charge collector, capturing and conducting the electrical energy due to its excellent conductivity. By integrating these two components in a crocheted structure, the design simulated a wearable cloth that combined the softness and flexibility of textiles with the functionality of smart electronics. This design provides a practical, textile-integrated solution for wearable energy harvesting, bridging the gap between electronic components and conventional textile fabrication techniques. The schematic and actual images of this crocheted prototype (D-CFC) are shown in Fig. 2(d), illustrating the adaptability of crochet techniques for producing wearable electronics.

Design 4.

The fourth prototyped design leverages the versatility of the functionalization process, which can be directly applied to

commercial textiles and garments. In this design, commercially available gloves (sourced from Decathlon™) were used as the substrate, demonstrating how existing textiles can be transformed into functional smart wearables in a cost-effective process. The functionalization process involved applying PPy polymer ink directly onto the surface of the gloves. This method offered the significant advantage of being highly adaptable to established manufacturing processes, making it easier to scale and integrate smart electronics into commercially produced textiles. The properties of the conjugated polymers were carefully tuned using doping and dedoping strategies. This allowed the polymer ink to be manipulated to achieve better conformity with the glove material, ensuring a good bond between the polymer and the textile. Additionally, the electrical properties of the polymer were optimized to meet the specific requirements of the energy-harvesting prototype. Once the gloves were functionalized to act as the active layer, CTFs were woven into the gloves, serving as the charge collector layer. The conductive fibers were integrated seamlessly with the gloves to capture the electrical charge generated by the functionalized polymer coating. This weaving process allowed for the creation of a fully functional wearable device, capable of energy harvesting while maintaining the comfort and wearability of standard gloves.

This design illustrates the potential for directly printing functional materials onto commercial textiles, making smart wearable technology more accessible and easier to incorporate into existing clothing. The images of these functionalized commercial gloves can be seen in Fig. 2(e), showcasing how everyday garments can be enhanced with electronic functionalities while preserving their original textile properties.

2.3. Electrical performance of the prototype devices

To explore the effect related to the number of fibers, three prototypes were built with different numbers of FTFs where all the fibers were functionalized for 30 min and all the devices were covered with PDMS: Devices with 3 EHF (D_{3f-30m}), 7 EHF (D_{7f-30m}), and 19 EHF (D_{19f-30m}). On the other hand, to study the effect related to different functionalization time (i.e., duration of polymerization), three prototypes were designed keeping the number (7 EHF) and length of each EHF unaltered: Devices were made with EHF functionalized for 30 min (D_{7f-30m}), 60 min (D_{7f-60m}), and 90 min (D_{7f-90m}).

Fig. 3(a) represents the instantaneous open-circuit voltage (V_{OC}) and short circuit current (I_{SC}) of devices D_{3f-30m}, D_{7f-30m}, and D_{19f-30m}, which recorded an average V_{OC} of 49.8, 125.6, and 244 V, respectively. The average I_{SC} was recorded to be 3.2, 8.5, and 15 μ A, respectively. Therefore, it can be deduced from the above results that there is a clear influence on the output when the number of EHF increases. Next, the influence of the duration of functionalization on the performance of the devices was investigated. Fig. 3(b) presents the electrical results of D_{7f-30m}, D_{7f-60m}, and D_{7f-90m}, which recorded the average V_{OC} of 125.6, 129.3, and 124.9 V along with the average I_{SC} of 8.5, 8.3, and 6.7 μ A, respectively. It can be concluded therefore that the average recorded output signals are slightly different depending on the functionalization time of PPy. From these results, we can see that D_{7f-30m} has shown comparatively higher average V_{OC} than D_{7f-60m} and D_{7f-90m}, even though no big influence was registered. These results corroborate the SEM and Raman analysis, which showed no significant difference between the polymer morphologies obtained in the synthesis with different reaction times. The PPy polymerization reaction is very fast, due to a high concentration of APS; therefore, even with a cooled down substrate and solution, it gives no significant electrical output difference with functionalization time variation.

The mechanical energy harvesting abilities of PPy can be described in 2 steps. (i) When pressure is applied onto the surface

of a PPy/electrode system, the polymeric networks will deform, resulting in a localized excessive rise in charge density, promoting Fermi-level pinning. Because of that, the charge translocation process occurs between the polymeric structure and the attached electrode system; consequently, an instantaneous current signal is generated while a load is connected to the outer system. (ii) Follows that, when the stress is released, the polymer networks are expanded, resulting in a localized charge scarcity (depinning of the local Fermi level). The neighboring regions, where the charge density is much higher, will balance out this scarcity by hopping mechanism, enabling a new pressure cycle to begin [62,63]. The schematic diagram for this mechanism is depicted in Fig. 3(c).

The real-life applications of any electrical device are associated with its power density and efficiency. A rectifier was added to the circuit to obtain a direct current (DC) output signal since the energy harvested directly from the devices is an alternate current (AC) signal. To calculate the power density of the devices, different external resistances were added to the rectifier circuit and the instantaneous output voltage of the system was recorded. The load added ranged from 55 k Ω to 150 M Ω (twenty-eight different loads). By having each voltage value associated with a different load, it is possible to calculate the power via $P = V^2/R$, and then each result is divided by the area to get the power density. The area considered in these calculations is the external surface area of the FTF that is coated with PPy. It is also important to note that the current density increases with the decrease of the external load in the circuit. For the D_{19f-30m} device, the maximum value recorded for power and current density was 2.29 W m⁻² and 19.5 mA m⁻² respectively, whereas for the D_{3f-30m} device recorded value was 0.23 W m⁻² and 23.9 mA m⁻², respectively. Compared to the D_{7f-30m} device, which shows a maximum power density (P_{max}) of 0.85 W m⁻² and current density of 22.4 mA m⁻², we can conclude that P_{max} increases with the number of EHF and that the maximum current density is approximately in the same range for the three prototypes, shown in Fig. 4(a). Fig. 4(b) shows P_{max} and current density for the D_{7f-60m} device of 0.52 W m⁻² and 14.2 mA m⁻², respectively, and that for the D_{7f-90m} device of 0.23 W m⁻² and 9.3 mA m⁻², respectively. These results illustrate a slightly higher performance of D_{7f-30m}. Thirty commercial blue LEDs (20 mA rating, 5 mm diameter, and 8.6 mm height) were also powered by instantaneous tapping (by hand), using the D_{7f-30m} device (Fig. 4d), without the need for a commercial capacitor. Moreover, 10 commercial blue LEDs were also powered by the D_{7f-60m} and D_{7f-90m} devices, separately. Using the D_{7f-30m}, fifty LEDs were powered by instantaneous patting as well. All the LEDs were connected in series. The video of device D_{7f-30m} lighting up 50 LEDs can be consulted in the [Supporting Information as Video S1](#).

In the pursuit of understanding the mechanical behavior of the devices, it becomes imperative to explore its performance across a spectrum of mechanical conditions. Previous mechanical tests have been conducted under specific and consistent parameters of frequency and force. However, it is essential to assess how the output responds to variations in mechanical conditions. To this end, an experimental setup was devised. This setup featured the utilization of an FSR406 force sensor, which was seamlessly integrated with an Arduino platform. This integration facilitated the real-time monitoring and acquisition of sensor data, thereby enabling the precise measurement of two key parameters: the instantaneous energy output (V_{OC}) and the force detected by the sensor, as depicted in Fig. 4(c). During the experimentation, there was deliberate variation of the mechanical conditions, specifically the applied force. The four selected force levels were categorized as follows: “very small force”, “low force”, “medium force”, and “high force”, corresponding to average values of 0.05, 0.7, 2.05, and 2.9 N, respectively. The corresponding V_{OC} output values obtained from these force inputs were observed to exhibit a direct correla-

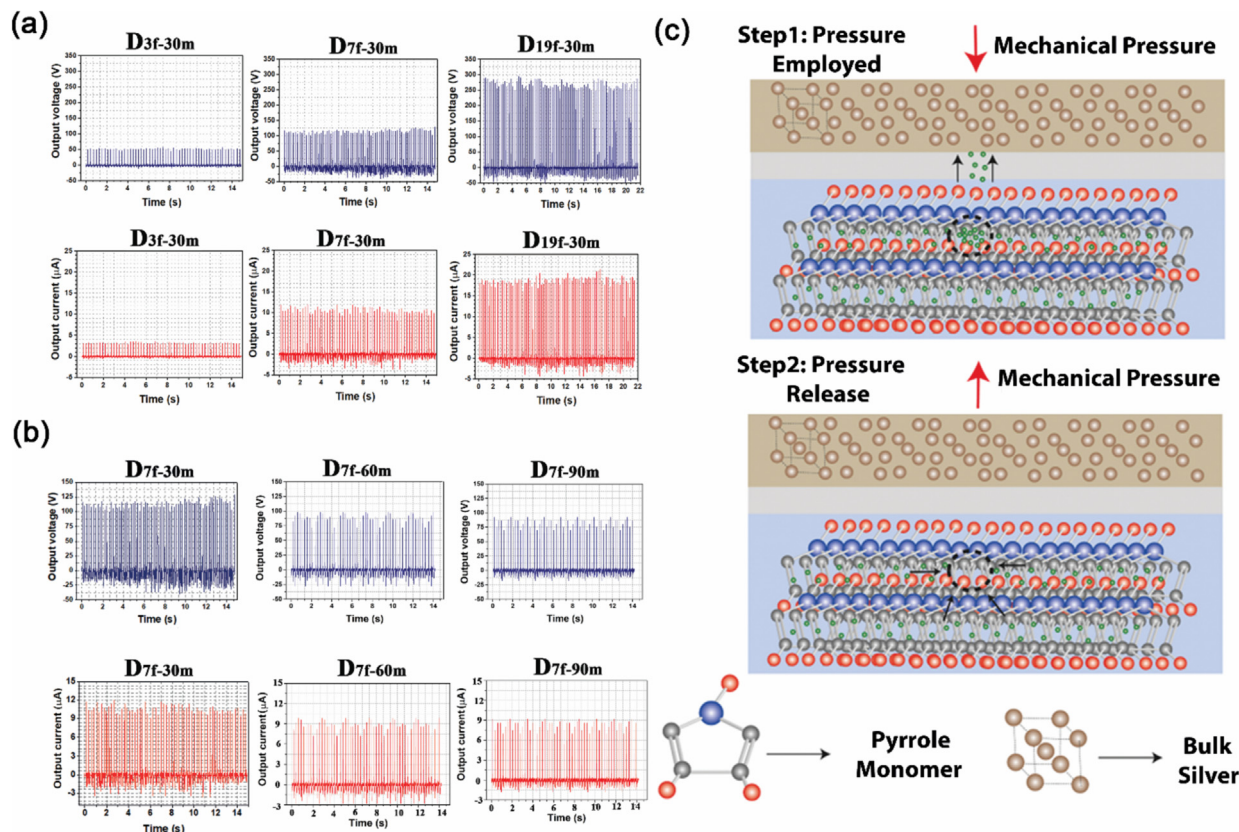


Fig. 3. (a) The graphics of the instantaneous V_{OC} and I_{SC} recorded for the control device D_{7f-30m} and for devices D_{3f-30m} and $D_{19f-30m}$, which portrays the influence of the number of EHF on the instantaneous output of the devices; (b) the graphics of V_{OC} and I_{SC} recorded for the control device D_{7f-30m} and for devices D_{7f-60m} and D_{7f-90m} , which portrays the influence of the functionalization time of PPy on the instantaneous output of the devices; (c) schematic design of the molecular mechanism that generates the transfer of charges from the polymeric structure to the charge collector layer of metallic nature. A localized pressure leads to an increase of localized charges, which results in a pinning of the Fermi level, and consequently translocation of charges. When pressure is released, the polymeric structure recovers and allows for a new pressure/release cycle.

tion with the applied force. Specifically, the average V_{OC} outputs for the aforementioned force categories were 2.89, 5.22, 20.61, and 37.38 V, respectively. This relationship underscores the significance of mechanical conditions on the performance of the devices.

Furthermore, to assess the stability and durability of the prototypes, an exhaustive mechanical stress test was conducted, involving more than 2000 cycles of mechanical loading. It is worth noting that while some slight discrepancies were observed in the results, the overarching trend remained consistent throughout the duration of the test. This consistency underscores the robustness of the devices under examination, as they maintained a similar performance trend despite the rigorous mechanical testing, thereby further validating the reliability of the experimental findings (Fig. 4e).

Electrical analysis was also done on the weaved prototypes. In terms of the V_{OC} and I_{SC} , the average output results of the D-MM prototypes are 19.5 V and 1.02 μA, respectively, and the values for D-MMC are 13.6 V and 0.62 μA, respectively (as shown in Fig. 5).

D-MM presented a maximum current density of 11.63 mA m⁻² and P_{max} of 41.3 mW m⁻², whereas D-MMC demonstrated a maximum current density of 2.16 mA m⁻² and P_{max} of 1.07 mW m⁻². Both devices reached a saturation value of 0.56 V while charging a 1 μF capacitor, device D-MM took 60 s, whereas D-MMC took 75 s. The presence of cotton fibers in D-MMC allows for fewer interface areas between the active layer and the charge collector layer, resulting in lower electrical performance. The energy stored can be calculated through the following equation.

$$E = \frac{CV^2}{2} \quad (1)$$

wherefrom we can derive that the energy accumulated in a 1 μF capacitor is 1.51×10^{-7} J. The relationship with power can be obtained through

$$P = \frac{E}{t} \quad (2)$$

wherefrom we can get the direct instantaneous power of the sensors, calculated to be 2×10^{-9} W for D-MM and 1.74×10^{-9} W for D-MMC. Hence, the functionalized weaved fabrics did not lose any significant electrical properties while incorporated into a cotton structure, which is quite good in this case, proving that our system has more potential for e-textile applications. Device D-CFC presented an average V_{OC} of 12.6 V and an average I_{SC} of 0.25 μA; furthermore it also demonstrated P_{max} value of 0.14 mW m⁻² and I_{max} of 0.154 mA m⁻². The general performance of device D-CFC was lower than that of D-MM and D-MMC, which may be due to the different electrode used. D-MM and D-MMC both use silver conductive fiber as electrode, whereas D-CFC uses carbon fiber. Silver has more electric affinity with PPy than carbon, since the Fermi level difference between conjugated polymers and metal electrode is lower in a PPy/Ag junction than that in a PPy/carbon junction. The same device reached a saturation level of 0.456 V in 75 s. Using Eqs. (1) and (2), we can derive the instantaneous energy and power of D-CFC, which was calculated to be 1×10^{-7} J and 1.38×10^{-9} W. Since device D-CFC has the lowest I_{SC} , the capacitor will reach saturation at a lower level, since the flow of charges is not enough to build more potential using a 1 μF capacitor. Table S1 compares all the devices and their outputs. The output performance obtained in the previous studies was measured by

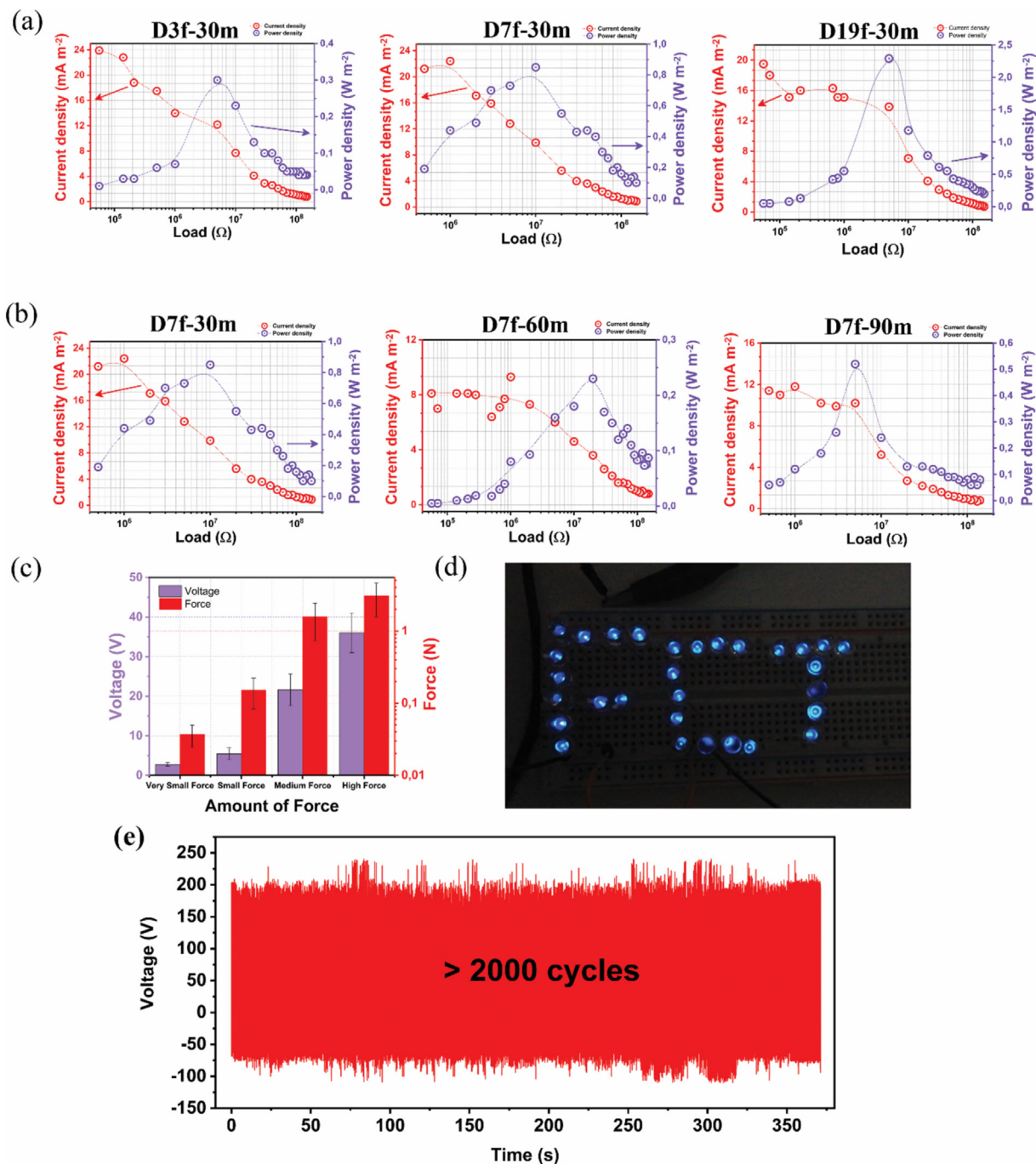


Fig. 4. (a) Power density and current density graphs for D3f-30m, D7f-30m, and D19f-30m devices; (b) power density and current density graphs for D7f-30m, D7f-60m, and D7f-90m; (c) mechanical tests conducted: FSR406 sensor was timed with V_{OC} from oscilloscope, averaging force values of 0.05, 0.7, 2.05, and 2.9 N and averaging V_{OC} of 2.89, 5.22, 20.61, and 37.38 V; (d) snapshot of LEDs lighted up during performance tests of D7f-30m; (e) mechanical endurance test under pressure for over 2000 cycles.

keeping similar magnitudes of force and frequency (force of ~3 N, frequency of ~6 Hz). In the field of flexible wearable electronics, the choice of active layer and electrode materials, as well as the structural design, plays a pivotal role in determining device performance and applicability. To elucidate the advancements in this domain, we compare our work with several recent studies in the literature [68–76] (Table 1).

Our work introduces a novel approach using silver fibers, cotton, and commercial textile fibers. These materials were selected to achieve exceptional electrical output of 244 V and 15 μA, resulting in current density 22.4 mA m⁻² and power density of 2.29 W m⁻². Notably, our design offers a unique combination of

wireless functionality, flexibility, lightweight construction, and long-lasting performance.

In comparison to existing studies, our approach stands out for its electrical performance and versatile applicability. While other works demonstrate varying degrees of electrical output and power density, our design excels in providing a robust solution for wearable electronics and motion monitoring applications. The integration of silver/PPy fibers with PDMS not only ensures high electrical performance but also enhances the durability and longevity of the device, making it suitable for prolonged usage in diverse environments.

Furthermore, the wireless capability of our design eliminates the need for cumbersome wiring, enhancing user comfort and mobility.

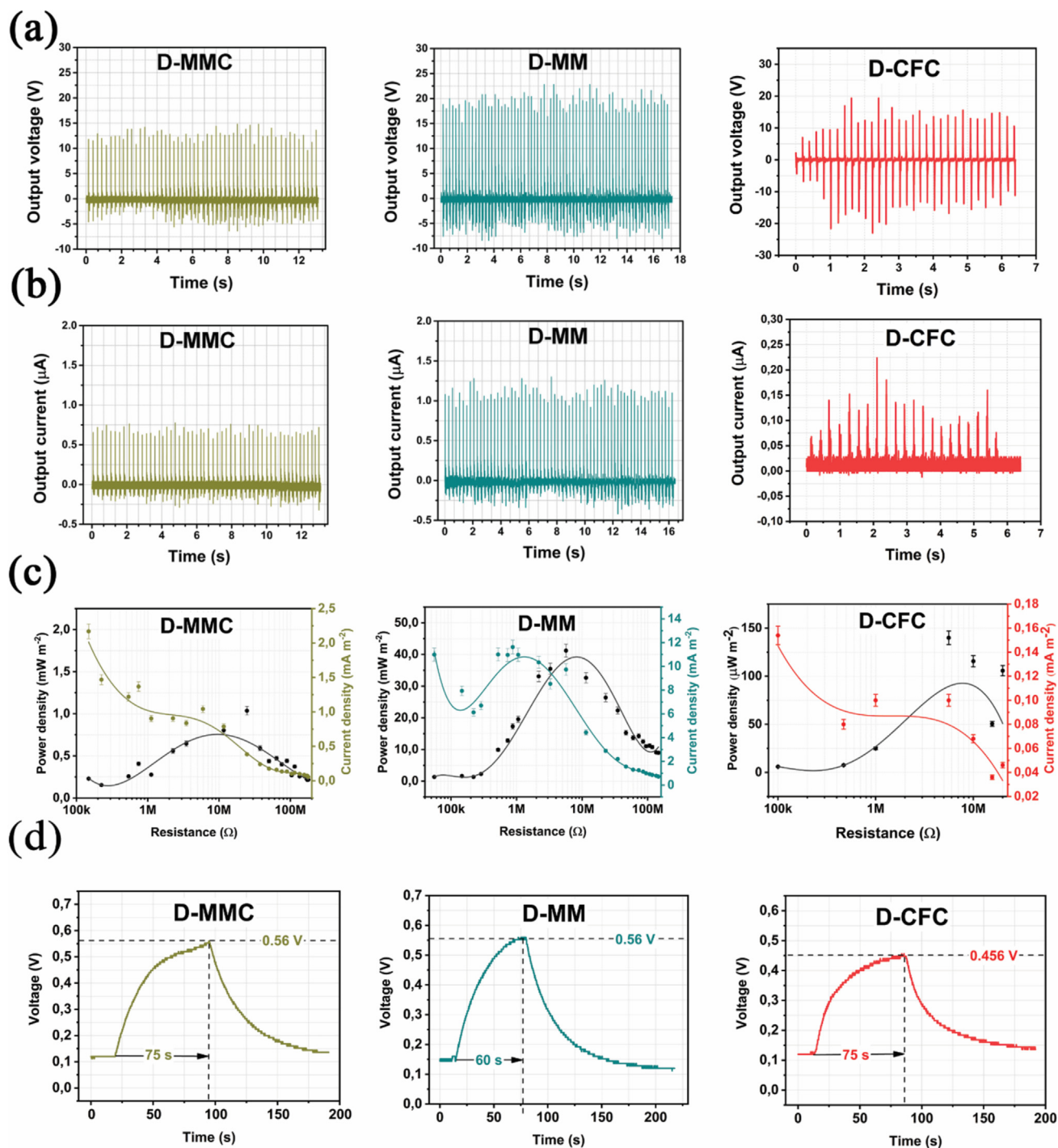


Fig. 5. The output characteristics of devices D-MMC, D-MM, and D-CFC. (a) V_{OC} obtained averaged 13.6, 19.5, and 12.6 V; (b) I_{SC} obtained averaged 0.62, 1.02, and 0.25 μA ; (c) P_{max} and I_{max} curves calculated and averaged $1.07 mW m^{-2}$ and $2.16 mA m^{-2}$ for D-MMC, $41.3 mW m^{-2}$ and $11.63 mA m^{-2}$ for D-MM, and $0.14 mW m^{-2}$ and $0.154 mA m^{-2}$ for D-CFC; (d) 1 μF charge capacitor test of D-MMC charged 0.56 V in 75 s, D-MM charged 0.56 V in 60 s, and D-CFC charged 0.456 V in 75 s.

This feature, coupled with the inherent flexibility and lightweight nature of the materials, makes our device particularly well-suited for seamless integration into clothing and accessories, offering users unprecedented freedom of movement without compromising performance.

2.4. Wireless gesture monitoring applications in wearables

One of the main focuses of this work is to exhibit the ability and/or prospects of the produced prototypes in real-life applica-

tions, for example in physical exercise monitoring applications. In order to test the sensing ability of the EHF, a simple bending test was made. Here, a PDMS encapsulation was made to protect and support just one EHF. The EHF with the PDMS encapsulation was attached to the finger (using lab gloves) and then was connected to an oscilloscope to check the instantaneous output voltage variation when opening and closing the hand. Fig. 6(a) shows the peaks formed upon bending of the fiber when the hand is closed. The test consists of bending the fiber at different angles to see how the sensing ability is affected by different types of

Table 1

Studies on energy harvesting using PPy functionalization for wearable applications.

Materials of active layer/electrode	Structures	Electrical output		Features	Applications	Ref.
		Voc/Isc	Power density			
HEC, pectine, chitosan/PPy	Film	273 V, 13 μ A	287 (μ W)	Flexible, antibacterial, mite removal	Wearable electronics and motion monitoring	[68]
BaTiO ₃ , PDMS, PPy/Al, PPy	Film containing nanoparticles	198 V, 18 mA m^{-2}	5.7 (mW)	Flexible, stretchable	Wearable electronics and pressure sensor	[69]
PVDF/PPy, Cu	Film	0.46 V	NI*	Flexible, piezo-photodetecting	Wearable electronics and self-powered vision	[70]
CsPbBr ₃ @PVDF/PPy	Nanofibers and beads	10.3 V, 1.29 μ A cm^{-2}	3.3 (μ W)	Flexible, temperature energy harvesting	Wearable electronics	[71]
Cotton fabric, PDA, CNT, PPy, HDTMS/Cu	Micro-nanorough fibers	180 V, 35 μ A	2.6 ($W m^{-2}$)	Flexible, humidity resistant, breathability	Motion monitoring and sensing as game blanket	[72]
PDMS, cotton/PPy, Cu	Fabric microfiber stacking	200 V, 6 μ A	82 (μ W m^{-2})	Flexible, easily manufactured	Wearable electronics	[73]
Carbon paper, PVDF/Cu	Micro-nano patterned film	20.2 V, 1.3 μ A	1.24 (μ W m^{-2})	Lightweight, flexible	Wearable electronics	[74]
Nylon fabric, cellulose, carbon nanotubes, PDMS/PPy fiber	Macrofibers	170 V, 0.8 μ A	54.1 ($mW m^{-2}$)	Biodegradable, super-strong, washable	Wearable electronics	[75]
Lycra fabric, chitosan, phytic acid/PPy	Microfibers	0.3 V, 1.8 μ A	7 (μ W cm^{-2})	Biodegradable, flexible, flame-retardant	Motion monitoring	[76]
Silver fibers, cotton, commercial textile, PDMS/Cu	Microfibers	244 V, 15 μA, 22.4 mA m^{-2}	2.29 ($W m^{-2}$)	Wireless, flexible, lightweight, long lasting	Wearable electronics and motion monitoring	This work

Hydroxyethyl cellulose (HEC), Polypyrrole (PPy), Polydimethylsiloxane (PDMS), Polydopamine (PDA), Polyvinylidene fluoride (PVDF), Carbon nanotubes (CNTs), Cetyltrimethoxysilane (HDTMS).

* NI: Not informed.

bending. For this reason, the angles of bending used in this test were 45°, 90°, and 180° (closed hand). The average result of the 45° of bending is around 495 mV, with a recorded maximum of 1 V. The average result of the 90° of bending is around 570 mV, with a recorded maximum of 1 V, as well. For the 180° of bending, the average result is 830 mV and the maximum peak recorded goes up to 2 V. This shows a very good sensitivity of the EHF to body movement when incorporated in wearable devices. Most importantly, this feature can be further developed for gesture monitoring systems.

A particular electronics strategy was employed. All these applications use an ESP32® platform to read the analog signals from the prototypes and translate them to digital signals. It is always our goal that is to employ these applications in the IoT paradigm. ESP32® is a microcontroller module with an embedded Wi-Fi and Bluetooth module. The ESP32® hardware was used to set up a hypertext transfer protocol (HTTP) communication system, to exchange data without connecting it to a World Wide Web protocol. The ESP32® can only read sensor values until 3.3 V; therefore a voltage divider is necessary to implement in the system, as well as

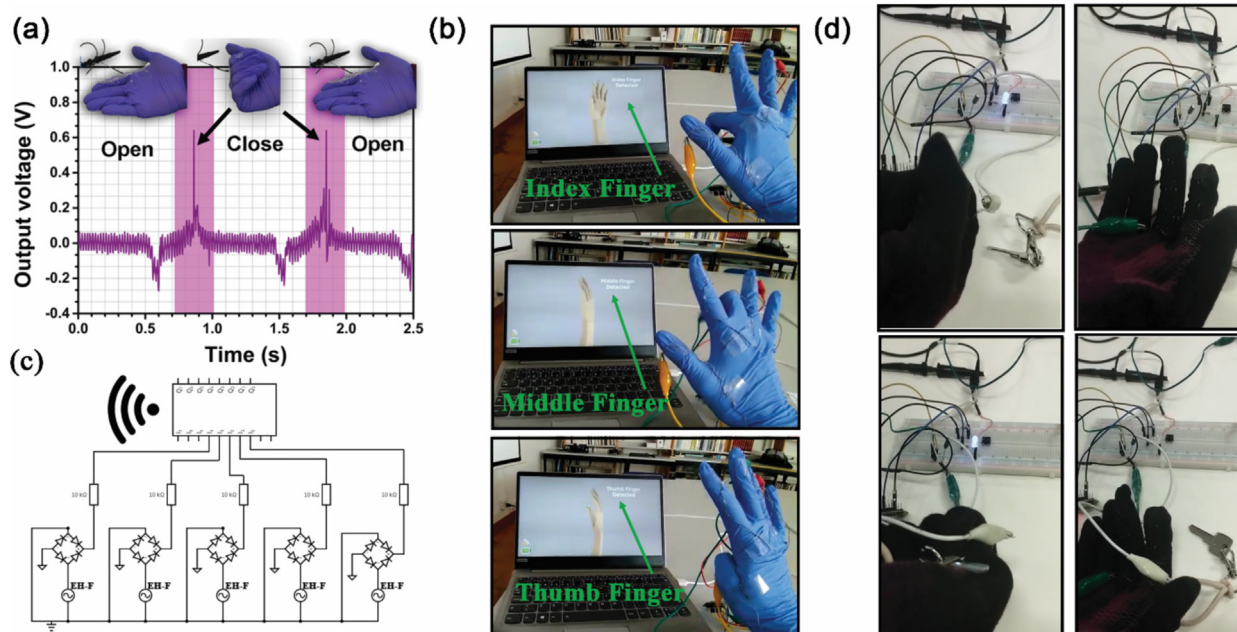


Fig. 6. (a) Sensitivity test on one single EHF to demonstrate its applicability in wearable IoT systems; (b) finger gesture monitoring system, designed with single EHF on each finger; (c) schematic of the general electronics strategy employed for the creation of gesture monitoring system; (d) gesture monitoring system, employed with functionalized commercial glove garment.

a rectifier to transform the AC characteristic of the mechanical energy harvesting devices into a DC characteristic. Finally, we used an external personal computer (PC) to create a Python program that would serve as a client, by receiving the digital information sent by the ESP32[®] and transcribing it to the desired interface (Fig. 6c). For the application presented in Fig. 6(b), a 3D animation of a moving hand has been designed, which connects the sensor readings from the movement of each finger to one EHF, which generates a pulse once the finger bends. This pulse is then detected by the system and the moved finger is identified. This application could be interesting as a proof of concept for the design of virtual reality (VR) interactive technologies, by designing the VR interactive cloths or gloves. Furthermore, we have also employed Design 4. in this system to simulate a gesture monitoring device made from commercial garment (Fig. 6d). The videos from Fig. 6(b) can be consulted in the [Supporting Information](#) as Video S2.

A gesture monitoring application was successfully put into practice. The devices were attached to different body parts (both

knee and elbow) of the users (Fig. 7a). With the users' movement, the device generates an electrical signal that is captured and translated to digital information by the ESP32[®]. The same module would then send the digital information to a web server. Another computer accessed said information and translated it to a digital image, representing the user's movement. A movie was recorded, displaying the working procedure of the system (Video S3); digital images of this application were also taken and are displayed in Fig. 7(b). A different protocol was implemented in the Python script, which allows to count several leg bending movements within a time-frame. As the user moves his leg, the counter displayed on the screen increases with the number of bending movements. Fig. 7(c) presents the snapshots taken during the experiment and the corresponding full video can be consulted at Video S4. In this experiment, one prototype of D_{19f-30m} was attached to the backside of the knee, covering the entire region of the popliteal fossa, and then it counted and displayed each full bending of the lateral collateral ligament.

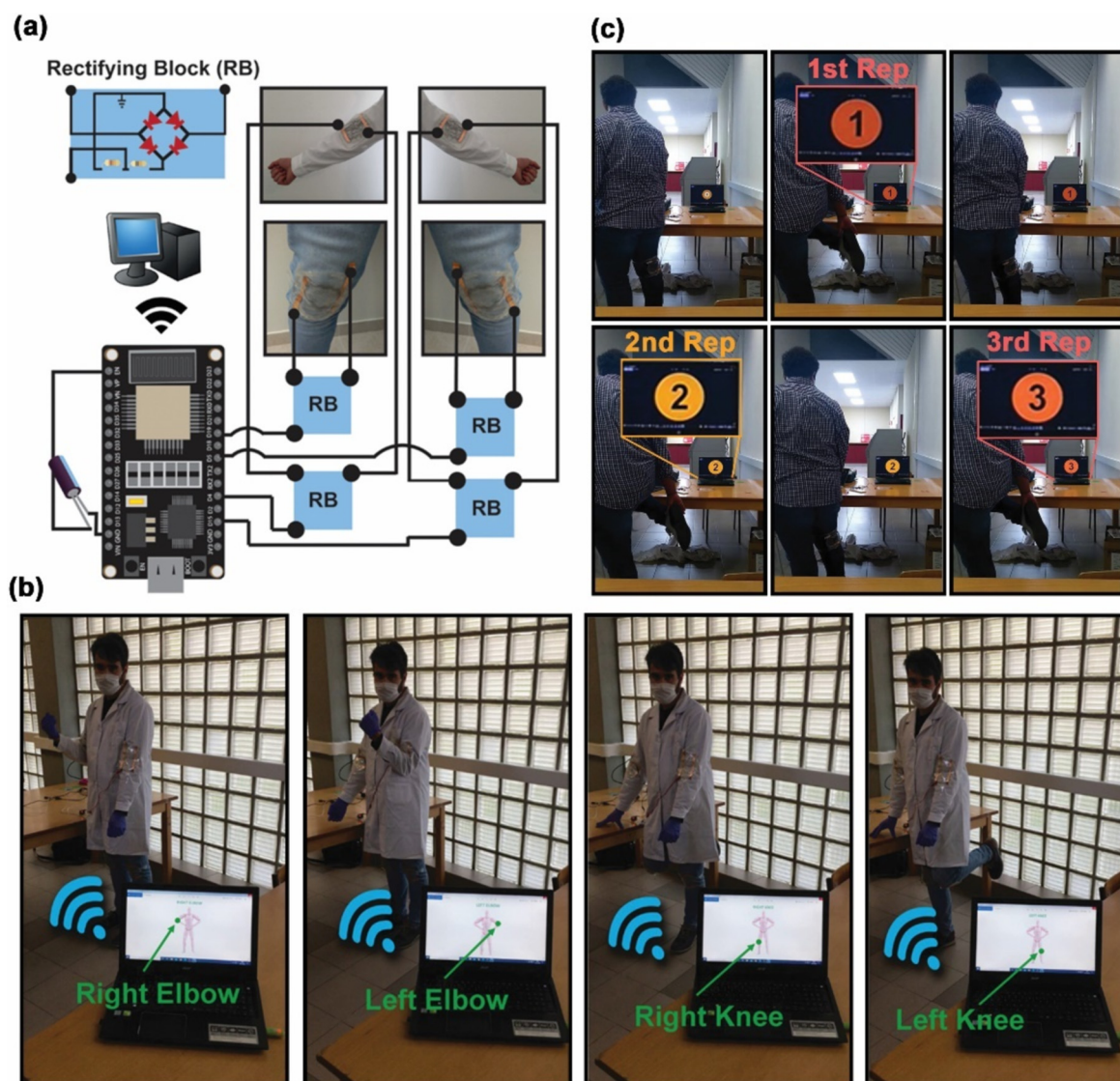


Fig. 7. (a) Schematic of the IoT sensing system comprising a rectifying block connected to each fiber sensor for physical monitoring applications; (b) movement monitoring application, where a sensor was attached to each limb, being able to identify which one had moved; (c) step analysis application allowing us to count the number of steps performed.

3. Conclusions

This work explores a novel approach to energy harvesting systems based on green materials and clean energy sources and uses them for health and physical exercise monitoring applications. Textile fiber and cloths are widely available materials in our day-to-day lives and are also trending in wearable electronics for human interactive applications. The great advantage of studying fibers/clothing materials is their low-cost, flexibility, and sustainable properties. During the present work, commercial silver-coated conductive textile fibers were functionalized with PPy and put together in two different ways: by PDMS casing and through weaving with a custom-built conventional looming frame. The wearable prototypes thus produced are sensitive to mechanical stress applied on top of it, generating an electrical output as a response. The main principle behind the operation of the device is the charge transfer mechanism that occurs between the π -conjugated polymer and the metal (electrode) layer when the system suffers from mechanical stress. These electrical signals are further utilized for gesture monitoring applications with a wireless block for IoT purposes.

Four different textile energy harvesting designs were employed. The best value of the power density and current density was 0.85 W m^{-2} and 22.4 mA m^{-2} , respectively, which correspond to the device made with only 19 CTFs functionalized in 30 min. The average instantaneous output voltage and current obtained for the said device were 244 V and 15 μA , respectively. These devices were also able to light up 50 commercial blue LEDs. All the devices have shown very good stability over time with slight variations over a period of 2000 cycles.

To test the implementation of these types of devices in real fabric applications, three prototypes were manufactured: D-MM (device not incorporated within cotton), D-MMC (device incorporated within cotton), and D-CFC (device with functionalized cotton and carbon fiber). Here, the best result recorded in terms of power and current density is from the D-MM device with 41.3 mW m^{-2} and 11.63 mA m^{-2} , respectively. The higher average V_{OC} and I_{SC} also corresponded to the D-MM device (19.5 V and 1.02 μA). The results indicate that the devices are stable enough for the e-textile application, as they were not affected due to handling during their weaving, enabling the production of conformable garments for smart textile applications.

However, the main intent was to design a self-powered gesture monitoring system, where the signals produced by the energy harvesting devices can be coded into digital signals and then translated as a movement monitoring application. When a body part is moved (e.g., arm), a signal is produced which is specifically translated so that the system identifies it as the arm. We have successfully programmed a system that can identify the specific body part that is being moved, which can be utilized for monitoring the body movement for medicine or sports purposes, e.g., by calculating the speed of movement or force being utilized while moving, as well as its potentiality as a gesture monitoring movement to be applied, for instances, in VR applications.

Experimental section

Experimental details can be found in the [Supporting Information](#)

CRediT authorship contribution statement

Guilherme Ferreira: Writing – review & editing, Writing – original draft, Software, Investigation, Formal analysis, Data curation. **Shubham Das:** Software, Formal analysis, Data curation. **Guil-**

herme Coelho: Software, Investigation, Formal analysis, Data curation. **Rafael R.A. Silva:** Writing – review & editing, Formal analysis, Data curation. **Sumita Goswami:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Rui N. Pereira:** Writing – review & editing, Supervision, Conceptualization. **Luís Pereira:** Writing – review & editing, Supervision, Resources, Project administration. **Elvira Fortunato:** Supervision, Resources, Project administration. **Rodrigo Martins:** Supervision, Resources, Project administration. **Suman Nandy:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jechem.2024.10.028>.

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