Since January 2020 Elsevier has created a COVID-19 resource centre with free information in English and Mandarin on the novel coronavirus COVID-19. The COVID-19 resource centre is hosted on Elsevier Connect, the company's public news and information website.

Elsevier hereby grants permission to make all its COVID-19-related research that is available on the COVID-19 resource centre - including this research content - immediately available in PubMed Central and other publicly funded repositories, such as the WHO COVID database with rights for unrestricted research re-use and analyses in any form or by any means with acknowledgement of the original source. These permissions are granted for free by Elsevier for as long as the COVID-19 resource centre remains active.
Single-arm diagnostic electrocardiography with printed graphene on wearable textiles

Ozberk Ozturk a, 1, Ata Golparvar a, b, 1, Gizem Acar a, Saygun Guler a, Murat Kaya Yapici a, c, d, * 

a Faculty of Engineering and Natural Sciences, Sabanci University, 34956 Istanbul, Turkey
b Integrated Circuit Laboratory, Ecole Polytechnique Federale de Lausanne (EPFL), 2002 Neuchatel, Switzerland
c Department of Electrical Engineering, University of Washington, 98195 Seattle, USA
d Sabanci University SUNUM Nanotechnology Research Center, 34956 Istanbul, Turkey

ARTICLE INFO
Keywords:
ECG
Single-arm electrocardiography
Heart-rate variability
Graphene textile
Electrocardiogram
Wearable electronics
Biopotential monitoring
Conductive nanomaterials
EHealth
E-textiles
Internet of things (IoT)
Long-term monitoring
Personalized healthcare
Reduced graphene oxide (rGO)
Smart garments
Vital signs

ABSTRACT
Stimulated by the COVID-19 outbreak, the global healthcare industry better acknowledges the necessity of innovating novel methods for remote healthcare monitoring and treating patients outside clinics. Here we report the development of two different types of graphene textile electrodes differentiated by the employed fabrication techniques (i.e., dip-coating and spray printing) and successful demonstration of ergonomic and truly wearable, single-arm diagnostic electrocardiography (SADE) using only 3 electrodes positioned on only 1 arm. The performance of the printed graphene e-textile wearable systems were benchmarked against the “gold standard” silver/silver chloride (Ag/AgCl) “wet” electrodes; achieving excellent correlation up to ~ 96% and ~ 98% in ECG recordings (15 s duration) acquired with graphene textiles fabricated by dip-coating and spray printing techniques, respectively. In addition, we successfully implemented automatic detection of heart rate of 8 volunteers (mean value: 74.4 bpm) during 5 min of static and dynamic daily activities and benchmarked their recordings with a standard fingertip photoplethysmography (PPG) device. Heart rate variability (HRV) was calculated, and the root mean successive square difference (rMMSD) metric was 30 ms during 5 min of recordings. Other cardiac parameters such as R-R interval, QRS complex duration, S-T segment duration, and T-wave duration were also detected and compared to typical chest ECG values.

1. Introduction

Wearable technologies (wearables) can facilitate effortless remote health monitoring in the comfort of the bedside and steer telemedicine to continuous mobile health tracking and medical assessment. Wearables in remote healthcare (mobile health, mHealth) expand rapidly in line with the expansion of “Healthcare 4.0” and the growing adoption of the internet of medical things (IoMT). Also, endorsement of machine learning (mL) and artificial intelligence (AI) algorithms with the merger of big data analysis in pathology, and application of 5G for securely and much higher probability of untimely death [6]. It goes without saying that wearable cardiac health tracker devices can provide an excellent convenience in the routine remote monitoring of people with cardiovascular complaints. Additionally, they can enable early diagnostics or set lifesaving alert mechanisms and inform first responders in case of cardiovascular complication events such as cardiac arrest [7]. Electrocardiography (ECG) is the conventional examination method to access vital information about the heart including rhythm abnormalities and various CVDs, through electrocardiograms (waveforms

* Corresponding author at: Faculty of Engineering and Natural Sciences, Sabanci University, 34956 Istanbul, Turkey.
E-mail address: murat.yapici@sabanciuniv.edu (M.K. Yapici).
1 Equal contribution

https://doi.org/10.1016/j.sna.2022.114058
Received 12 September 2022; Received in revised form 4 November 2022; Accepted 23 November 2022
Available online 24 November 2022
0924-4247/ © 2022 Elsevier B.V. All rights reserved.
representing electrical activity of the heart) which are picked up while the heart muscle fibers contract in response to electrical depolarization. These electrical activities (cardiac biopotentials) are commonly recorded by placing electrodes on the chest and limbs [8]. Another competing technology for recording cardiac information is photoplethysmography (PPG). Although PPG devices are inherently optimal for mobile health monitoring and can be integrated even to an earbud or a ring [9,10], the amount of extracted cardiac features from PPG is not comparable to the rich ECG data.

However, one of the inherent challenges of mobile ECG recording is optimizing the number of channels and corresponding electrode placement since conventional clinical ECG involves a complex 12-lead monitoring mechanism and hinders its mobile function. Newly introduced, smartwatch ECG approaches use a derivative of typical lead-I ECG and are adequate for the mobility specifications, but they best perform in rest conditions and lack continuous monitoring capability due to their electrode arrangement [11]. On the other hand, wrist ECG addresses ongoing monitoring issues of smartwatch ECG, but they are still not optimal for easy integration to everyday wearable clothes like smartwatch ECG devices [12]. Ear-ECG and neck ECG are other alternatives, which seems to address both continuous monitoring and easy integration to daily clothing challenges, but either they have weaker signal due to distance from the heart [13,14], or are more affected by motion artifacts due to their arrangement [12].

Another possibility is exploiting single-arm ECG. Not long ago, a study looked into finite element method simulation of ECG over body surface by constructing the cardiac current dipole by superimposing three orthogonal current dipoles with the amplitude variations in each dimension and simulated the actual electrical behavior of the heart [15]. It illustrated by simulation that the intensity of the R-wave peak is more intense in the left arm compared to any other non-standard ECG electrode configuration on the upper torso and head surface. Following this insight, we present measurement results and a biopotential map confirming that upper left arm region below the shoulder deltoid yields the highest R-wave amplitudes (Fig. 1). Thus, left arm ECG’s arrangement advances are two-fold; first, ECG is more intense in this spot than ear-ECG and others; second, motion artifacts less affect the signal than neck-ECG. Both advances promise better signal-to-noise ratio (SNR) measurement and render extraction of more cardiac parameters such as event durations of R-R interval, QRS complex, S-T segment, and T-wave from ECG data, which was not recorded in high accuracy with other non-standard ECG arrangements.

On the other hand, upon choosing electrodes’ arrangement, selecting sensing electrodes’ type is the next design challenge for a mobile ECG monitoring device. Due to their stable characteristics, silver/silver chloride (Ag/AgCl) “wet” electrodes supported by adhesive backrest and gel layer, to improve skin-electrode contact, are widely used in clinical ECG [16]. However, the conductive gel dehydrates in time and degrades the electrode’s SNR performance. Therefore, electrodes need to be replaced in a few hours or gel needs to be re-applied, which are neither practicable in everyday use nor easy to integrate into any mobile ECG solution. Moreover, the gel can cause an itching sensation and develop red and swollen skin immediately upon peeling the electrode. Such irritations and allergic reactions may last for several hours [17] or, in certain conditions, are reported to lead to dermatitis [18]. Therefore, it is preferable to use gel-free “dry” electrodes in continuous monitoring applications due to their superior long-term and multiple-day measurement performance [19]. Compared to other variants of dry electrodes, electronic textiles (e-textiles) appear to be the key technology in wearables non-invasive monitoring of biological indicators [20]—other competing technologies include tattoo-based and bio fluidic-based wearables [21,22].

E-textiles promise flexibility, permeability to air and moisture, and accessible integration to daily clothing. Flexibility is essential to enable skin-compatible devices by matching the body’s natural contours, providing wearability, and achieving better skin-electrode coupling, whereas permeability to air and moisture alleviates the possibility of skin irritations [17]. Moreover, intelligent clothing—clothes enhanced with e-textiles to add functionality beyond traditional use, can embrace a critical role in a more extensive wireless body area network (WBAN) by including embedded flexible and stretchable antennas [23] and wireless transmission modules to enable continuous transfer of physiological information, which had sensed by the fabric itself, to a remote medical unit or the cloud; to be examined on the spot or later by professional healthcare staff. On the other hand, the flexible interconnection of the sensing sections of the intelligent clothes with electronic circuits is possible [24]. Additionally, in order to have truly “all-textile” intelligent clothes, research is actively progressing on the development of textile-based lithium-ion batteries [25] and textile-based energy harvesting mechanisms [26].

Fabrication techniques in textile electrodes also play a critical role in expanding intelligent clothing. Textile electrodes can be attached to daily clothes and accessories thanks to instream fabric manufacturing/ decoration approaches such as knitting, weaving, and embroidery [27]. On the other hand, the sensing electrode section can also be applied onto finished textiles directly using techniques such as electroplating [28], physical vapor deposition (PVD) [29], chemical polymerization [30], dip-coating [31], or printing methods [32]. Especially, dip-coating and spray printing are promising since low manufacturing cost due to the adaptability of the existing commercial roll-to-roll (R2R) textile fabrication processes presents a serious game changer over the other e-textile fabrication methods, which usually require special equipment or processes that hinder their mass production (e.g., deposition functional thin films or patterning with lithography-based techniques in a cleanroom environment) [33,34].

Graphene-based nanocomposites are emerging as attractive materials for biomedical applications, and various highly flexible and stretchable sensors have been fabricated using graphene for wearable technologies, including flexible strain gauges and electrochemical biosensors [35,36]. Our group has been pioneering the synthesis of graphene e-textiles for biopotential signal monitoring, and we have introduced a cost- and time-effective scalable process based on dip-coating whereby conductive textiles are formed using graphene as a cladding layer [31]. Furthermore, previously, we benchmarked the

---

**Fig. 1.** Measured R-peak amplitudes recorded from different locations across the left arm displayed as a biopotential heat map.
2. Materials and methods

2.1. Fabrication of graphene textiles by dip-coating and spray printing

Graphene, a 2D single-layer structure of sp² bonded carbon atoms formed in a honeycomb pattern, has recently attracted significant attention due to its unique electrical and structural properties [43]. Graphene-coated conductive textiles were prepared following a scalable and inexpensive, three-step “dip-dry-reduce” process to obtain conformal claddings of graphene over textile fibers. The fabrication process started with synthesis of graphene oxide (GO) following Hummer’s method and applying the GO solution (4 mg GO dispersed in mL of distilled water) onto textile surface using dip-coating and spray printing techniques. The dip-coating was applied by immersing the previously cut plain textile in GO solution with 3 × 3 cm² dimension (Fig. 2).

Then, each coated piece was dried to achieve homogeneous GO-coated textiles. The surface texture, tightness, and thickness are essential factors in textile coating; these parameters were controlled during the process. Various textiles such as nylon, polyester, and cotton have been tested to obtain optimal deposition. Due to the low surface roughness, which resulted in homogeneous GO coating, nylon is selected as a plain textile to provide the best coating uniformity.

In parallel, the spray printing technique was engineered to pattern the graphene textile coatings, which requires predetermined electrode positioning. These positions were selected through trial-and-error ECG experiments with the dip-coated electrodes on single-arm recordings. Then, stencil and textile were placed on a hot plate to accelerate the drying of the textile. The spray was held at ~ 30 cm distance from the textile during the printing of GO and was followed by the “print-dry-reduce” fabrication cycle (Fig. 2). Next, chemical reduction was performed to create conformal electroconductive graphene structures using 4.33 mL of a 10 vol% saturated sodium hydroxide (NaOH) buffer solution and 227 mg of sodium borohydride (NaBH₄) as a reducing agent following the process described by Guex [44]. The chemical reduction process disrupts the sp² bond of graphite, making the epoxide and hydroxyl groups hydrophilic [45,46]. The quality of the properties and potential application areas of the fabricated graphene may vary according to the selected reduction process [47].

After reduction, textiles were washed by immersing them in deionized water to remove unbounded chemical residues from the fiber surface, and only stable and well-adhered reduced graphene oxide (rGO) flakes remained on the textiles. Finally, they were placed in a vacuum oven and left to dry for a couple of hours. During the reduction of GO, there was a noticeable change in the textile color from brown to black due to the removal of oxygen-containing groups, which accompanies the increase of the electrical conductivity. Thus, the color change of the textile is a qualitative indicator of the effectiveness of the reduction process.

For prototyping, the prepared e-textile piece from the dip-coating
procedure was fixed on a flexible sticky foam and sandwiched on an ordinary sports headband between metallic snap fasteners to establish electrical connection with the front-end circuitry. To acquire higher SNR and guarantee skin-electrode interface stability, we have used elastic bands with Velcro straps and polyethylene-based foam paddings to provide pressures in the range of a few mmHg (up to 5 mmHg) to support the contact of graphene-based e-textile sensors and form a stable high contact area on the skin surface. The procedure for printed textiles was more straightforward, and they required no cutting since graphene was homogeneously distributed on all its surfaces, including the edges.

In the dip-coating, the color of the edges was slightly darker compared to the central spots, indicating looser homogeneity distribution of the coating. In contrast, spray printing enables denser coating of the textile surface with the transfer of liquid droplets using pressurized air as opposed to bulk liquid transfer in the dip coating. Moreover, spray coating also allows for precise adjustment of the electrode locations in the final cloth as this approach enables patterning of the functional graphene layer onto a larger fabric. To expand the spray printing technique to accommodate additional electrodes and recording channels, the stencil (template) can be redesigned and produced using 3D printing or injection molding, making this approach adaptable for rapidly configuring and fabricating smart textiles on demand.

2.2. Scanning electron microscopy imaging and Raman spectroscopy on graphene textiles

Graphene textiles were imaged with a scanning electron microscope (SEM) and Raman spectroscopy was performed as a means to quickly confirm the coating and GO reduction steps. As illustrated in the scanning electron micrographs (Fig. 3a), surfaces of the textile fibers show high contrast due to fairly homogenous layering of rGO while the electrically insulating cores of the fibers are bright and display charging. This is one visual indication of the existence of a conductive rGO coating on textile surfaces. Along with a visual confirmation, the changes in chemical structure during the reduction process of GO can be observed with Raman spectroscopy. Typically, crystalline graphite shows only a sharp Raman shift called the G-peak in the 1575–1581 cm$^{-1}$ region, while GO and rGO display two broader Raman shifts named as the D- and G-band (I$_D$/I$_G$) occurring at slightly different locations and with different intensities [48,49]. Reports in the literature have extensively studied the Raman spectrum of GO and rGO obtained with different synthesis and reduction chemistries, and while there may be slight differences in the D and G vibration bands, typically for GO the D-band can peak in the range of 1353–1363 cm$^{-1}$ and G-band in the 1588–1598 cm$^{-1}$ range; while for rGO, the D-band can peak in the range of 1349–1352 cm$^{-1}$ and G-band as low as 1574 cm$^{-1}$ and up to 1603 cm$^{-1}$ range have been observed [49–51]. To verify the existence of the typical D and G vibration bands in the synthesized GO and rGO textiles, we have performed Raman spectroscopy (Renshaw, England, UK) using 532 nm laser excitation. The recorded Raman spectrum of GO and rGO textiles (Fig. 3b) display D and G vibration bands well aligned with the literature. Moreover, the ratio of the D and G peak intensities (I$_D$/I$_G$ ratio) is used as a typical signature of GO to rGO conversion, whereby, an increase in the I$_D$/I$_G$ ratio is expected upon reduction [52,53]. While the I$_D$/I$_G$ ratio was 0.89 for GO coating on textile, it increased to 1.38 upon chemical treatment of the GO-coated textile with NaBH$_4$, which validates the reduction process and formation of reduced graphene oxide (rGO) on textile surfaces.

2.3. Skin-electrode impedance, sheet resistance, conductivity and washability

The skin-electrode impedance is a prominent performance metric for biopotential electrodes, where small and stable impedance values indicate better performance [54,55]. The impedance of the graphene textile electrodes and the Ag/AgCl electrodes were compared. The three electrodes were placed on the forearm, ~5 cm away from the wrist. The produced current was applied to the skin by the placed electrodes, and the reference electrode was held at ground potential. The measuring electrode was either an Ag/AgCl or textile electrode. The impedance of the Ag/AgCl electrode in the frequency range of 1 Hz to 1 kHz was measured between 74 kΩ and 16 kΩ, while the impedance value of the graphene-coated textile electrode ranged from 67 kΩ to 16 kΩ (Fig. 4).

Lower impedance values for the graphene textile electrodes were attributed to having a larger area than the Ag/AgCl electrodes, 9 cm$^2$ vs. 2.3 cm$^2$ respectively, due to the inverse linear relationship between electrode area and skin-electrode impedance. Impedance values are affected by the electrode surfaces that are in contact with the skin, and since textile electrodes do not incorporate adhesives, their surface contact is not excellent, and the physical electrode area does not directly translate to the active electrode area.

In addition, the sheet resistance of graphene textiles was measured with a four-point probe unit. The sheet resistance and conductivity were found as ~14 kΩ/square and ~0.33 S/m, respectively, for a textile thickness of ~0.2 mm (Fig. 4b). The developed graphene textile electrodes are highly flexible due to their soft, fabric nature and only weighing ~50 mg.

One of the characteristics required for the long-term usability of textile electrodes is to assess their level of degradation to washing. Indeed, loss in conductivity is expected after a certain number of washing cycles due to absorption of water and mechanical stress load on textile fibers which may decrease or damage the conductive network. Starting with two identical electrodes, one was cleaned in a washing machine (40 °C at 400 rpm for 30 min), and the other was gently dipped to detergent water and kept without movement for 30 min (hand
It was found that after five washing cycles, hand washed electrode’s resistivity increased 50%. In contrast, the resistivity of the other electrode increased by 150% due to the high mechanical stress induced by the washing machine, indicating hand wash should be preferred for graphene textiles to preserve their long-term usage. Additionally, surface resistance increased from \( \sim 45 \, \text{k\(\Omega\)} \) to \( \sim 65 \, \text{k\(\Omega\)} \) in five handwashing cycles. However, considering the overall circuit model of the skin-electrode (with impedance \( Z_E \)) and analog front-end (input impedance \( Z_{IN} \)) as two impedances in series, the washing cycles did not prevent reusage of the electrodes for ECG measurements, due to much higher input impedance of the analog front-end amplifier compared to the skin-electrode impedance, and the gain provided by the amplifier [17].

### 2.4. Design of the front-end readout circuitry

It is imperative to condition the signal and filter the motion artifacts and power-line noise to record ECG signals with an excellent SNR. Overview of the complete, wearable single arm ECG monitoring and acquisition system with printed graphene electrodes and embedded electronics is shown in Fig. 5, along with the schematic of the custom-designed readout unit. The values of circuit components are summarized in Table 1. At the analog front-end, the circuit obtains its surface potentials via graphene textile electrodes, and after onboard conditioning, they are sent to the microcontroller (MCU) for digitization and further processing. Then, signals are transmitted via a Bluetooth connection (HC06) to the PC with a custom-developed graphical user interface in LabVIEW (National Instruments, USA), where real-time data is stored and tracked.

The onboard analog circuitry has second- and fourth-order Sallen-Key Butterworth high-pass and low-pass filters with 2 Hz and 39 Hz cut-off frequencies, respectively. The values for the filters were optimized and simulated using Texas Instrument’s online filter design tool. The instrumentation amplifiers (INA122, Texas Instruments, USA) and op-amps (OPA2365, Texas Instruments, USA) suit the battery operation with a single lithium-ion polymer battery (3.7 V and 500 mAh). A voltage divider (MAX5421, Maxim, USA) was used to adjust the gain in the post-amplification level. MCP73831 (Microchip, USA) and TPS61090 (Texas Instruments, USA) were used in the circuitry to add battery charging capability and DC-DC boosting. A rail-splitter (TLE2426, Texas Instruments, USA) with a voltage divider with a buffer circuit was applied to avoid unbalanced occurrences when dividing the regulated 5 V. The entire system was placed in a 6.5 x 5 cm enclosure.

![Diagram of the ECG monitoring system](image)

**Table 1** List of components and specifications for the signal acquisition system.

| Circuit Blocks        | Specifications                  |
|-----------------------|---------------------------------|
| Pre-amplifier         | \( R_{1}, R_{2}: 1 \, \text{k\(\Omega\)}, C_{1}, C_{2}: 1 \, \text{nF}, C_{3}: 100\, \text{nF} \) |
| Protection circuitry  | \( R_{2}, R_{3}: 20 \, \text{k\(\Omega\)} \) |
| DRL circuitry         | \( R_{2}: 10 \, \text{k\(\Omega\)}, R_{1}: 390 \, \text{k\(\Omega\)}, C_{1}: 1 \, \text{\mu F}, C_{2}: 270\, \text{nF} \) |
| High-Pass Filter      | \( R_{1}, R_{2}: 36 \, \text{k\(\Omega\)}, R_{3}: 4.7 \, \text{k\(\Omega\)}, R_{4}: 1 \, \text{k\(\Omega\)}, C_{5}, C_{6}: 1 \, \text{\mu F}, C_{7}: 1 \, \text{nF} \) |
| Low-Pass Filter       | \( R_{12}, R_{13}: 10 \, \text{k\(\Omega\)}, R_{14}: 160 \, \text{\Omega}, R_{15}: 330 \, \text{k\(\Omega\)} \) |
| Battery               | Li-ion battery                  |
| ADC                   | MCP73831                        |
| Bluetooth Module      | TPS61090                        |
| Power Management      | TLE2426                          |

![Diagram of the system components](image)
2.75 cm³ transparent case and it weights ~ 53 g.

3. Results and discussion

3.1. Single-arm ECG with dip-coated graphene textiles

In order to verify the feasibility of the developed graphene textile electrodes with dip-coating in sensing single-arm electrocardiograms, they were benchmarked against the standard AgAgCl electrodes (Foam Monitoring Electrodes Ref 2228 3M Red Dot, USA). ECG recording experiments were performed in static and dynamic conditions for 5 min each, where the subjects were instructed to sit in a relaxed position and walk, respectively. Due to the importance of the electrode position on the signal quality, signals were asynchronously recorded so that the comparisons could be from the exact electrode locations (first with armbands, then wet electrodes). There were 8 participants (3 female, 5 male), and none reported a CVD history. Since the measurement was done asynchronously, the heartbeats and, therefore, the P-QRS-T complex were naturally not synchronous in recordings of different electrode types. Thus, we selected a 15 s spot with matching heartrates from each recording and synchronized them in data processing for obtaining correlation values. To quantify the overlap between the optioned signals, the built-in linear correlation function of MATLAB® (Mathworks, USA) was used. The correlation coefficients summarized in Table 2 reveal a maximum correlation of 96% obtained ECG recordings from participant 1 (P1) and minimum of 84% observed in participant 8 (P8) with an overall average of 88.6% in the static trials, and a maximum of 93% for P1 and minimum of 70% for P3 with an overall average of 84% in the dynamic trials.

Fig. 6 illustrates the results from participant #1 who had the highest correlation coefficient on both trials, and each segment of P-QRS-T morphology is observed both in static and dynamic trials in single-arm ECG recordings obtained with AgAgCl electrodes and dip-coated graphene textiles. During the walking trial, P-wave was the most challenging feature to observe from all the participants. This is attributed to the nature of single-arm ECG measurement approach, which results in its fluctuation [56]. It was observed that signals taken from graphene electrodes had slightly better skin-electrode impedance values (P- QRS-T morphology) from the noise (baseline and its fluctuation) [56]. It was observed that signals taken from graphene textiles were slightly better than AgAgCl electrodes (Table 2) since graphene electrodes had slightly better skin-electrode impedance values in low frequencies. As SNR values inherently differ from person to person due to physiological variations, a fair SNR assessment would be to compare the different electrodes and trials for the same participant as opposed to comparing the averaged values of all the participants. This is fundamentally due to the ECG intensity difference per person (especially R-wave peak intensity); that is, if the ECG signal’s overall intensity is more than the baseline, it will have high SNR.

Factors such as fitness and age have a role in achieving high SNR from ECG signals. For instance, if recording happens to be picked up from individuals with more fat tissue on their arm, due to the low-pass filtering of these tissues, surface electrodes will pick lower ECG signal intensity than more slim participants, and their SNR values will be lower. However, this does not mean their correlation coefficient is low [57]. Likewise, individuals with body hair on arms also record less SNR intensity with textile and clinical electrodes. For instance, participant 1 had the highest correlation value, but his SNR values were below average. As discussed, this is attributed to the quality of the ECG signal picked up irrespective of the type of electrodes.

3.2. Single-arm ECG with spray-printed graphene textiles

Right after experimentation with the first prototype based on dip-coated graphene textiles, participants 7 and 8 who were among the “poor” performers in terms of both correlation and SNR values, were selected to test the spray-printed graphene textiles and ECG signals were recorded using the same experimental protocols. As previously mentioned, three graphene textile electrodes separated roughly ~ 10 cm apart were directly printed onto a bare nylon textile surface, unlike the dip-coating approach where the textile electrodes were sewn onto a piece of nylon textile which acted as a “carrier”. Fig. 7 shows sample of a recorded ECG signal from participant 8 under static and dynamic conditions for a duration of 15 s with a clear display of the P-QRS-T complex, as well as, a longer recorded signal of more than 1 min where R-peaks are visible.

The correlation coefficients among signals recorded with two different electrodes (spray-printed graphene textiles and “wet” AgAgCl electrodes) along with SNR values for each participant under static and dynamic conditions are listed in Table 2 as P1’/ P2’. Results show that, on average, the second prototype based on spray-printed graphene

Table 2

| Participant (P) No / Age | Correlation [%] | SNR Sitting [dB] | SNR Walking [dB] |
|------------------------|-----------------|------------------|------------------|
|                        | Sitting         | Walking         | AgAgCl           | Dip-coated Graphene Textile | AgAgCl           | Dip-coated Graphene Textile |
| P1 / 26                | 96%            | 93%             | 13.7             | 13.3             | 14.1             | 12.3             |
| P2 / 22                | 93%            | 85%             | 25.9             | 24.1             | 16.0             | 20.2             |
| P3 / 45                | 85%            | 70%             | 22.0             | 21.9             | 21.4             | 20.2             |
| P4 / 31                | 90%            | 89%             | 23.0             | 24.1             | 18.9             | 19.4             |
| P5 / 24                | 87%            | 86%             | 24.1             | 21.4             | 15.7             | 18.6             |
| P6 / 52                | 87%            | 74%             | 20.7             | 22.6             | 18.0             | 23.7             |
| P7 / 27                | 87%            | 89%             | 15.1             | 13.8             | 15.0             | 15.9             |
| P8 / 26                | 84%            | 86%             | 18.3             | 23.3             | 18.1             | 23.1             |
| Mean: 31.6             | 88.6%          | 84%             | 20.4             | 20.6             | 17.2             | 19.2             |
|                        | Sitting         | Walking         | AgAgCl           | Spray-coated Graphene Textile | AgAgCl           | Spray-coated Graphene Textile |
| P1’ / 27               | 93%            | 84%             | 25.8             | 27.5             | 22.0             | 22.2             |
| P2’ / 26               | 95%            | 92%             | 26.9             | 30.6             | 22.1             | 25.0             |
| Mean: 26.5             | 94%            | 88%             | 26.3             | 29.1             | 22.1             | 23.6             |
Fig. 6. Single-arm ECG of a participant with the highest correlation coefficient in the static and dynamic condition, recorded by the dip-coated graphene textile electrodes.

Fig. 7. Single-arm ECG with spray printed-based prototype in the static and dynamic experiments. Compared to the dip-coated-based prototype, correlation coefficients are high even in longer durations, thanks to the superior coating.
3.3. Real-time detection of heartrate (BPM) and heart-rate variability (HRV)

In parallel with the real-time data acquisition, the system detects critical ECG parameters such as R-R interval, heartrate (BPM), and heart-rate variability (HRV) and reports them to the operator in real-time. HRV indicates the change in the intervals of the consecutive heartbeats and is a critical index for monitoring the cardiovascular system and routinely used for detecting different types of arrhythmia [58]. HRV is also associated with physical fitness and psychological well-being and is used in recovery and stress analysis [59]. During the COVID-19 global pandemic, the HRV data is heavily analyzed for early disease diagnosis [60], and also to study the effect of isolation on mental health and the stress levels of individuals [61].

R-wave peaks were detected with a thresholding-based algorithm in which a single-arm ECG signal was smoothed twice with a moving root mean square (RMS) filter with a 150 ms window. The algorithm calculates the threshold parameter with an automated calibration upon starting the recording. In every 2-minutes of R-wave peak detection, approximately two T-waves were mistakenly overcounted as R-peaks, which is still an acceptable error rate for a simple thresholding-based heartrate detection algorithm. After calculating the R-R interval, root mean square successive difference (rMSSD) was calculated, which is one of the popular time-domain detection metrics for HRV [62]. For the previously mentioned 5-minute trial, the rMSSD-HRV value was calculated as 30 ms, which lies inside the typical expected range (i.e., 18–74 ms) for a 30-year-old participant.

Since the developed system can capture the intricate features of the P-QRS-T complex with an acceptable SNR and sufficient intensity (Fig. 8a); information on other critical cardiac parameters can be

| Parameter | Single-arm ECG | Single-arm ECG | Typical chest ECG |
|-----------|----------------|----------------|-------------------|
| System    | "Custom system" | "Commercial system" | "Clinical system" |
| BPM sitting [count] | 74.4 ± 8.6 | 72.4 ± 6.2 | 60 – 100 [63] |
| BPM standing [count] | 87.3 ± 8.7 | 76.2 ± 6.6 | 70 – 110 [63] |
| BPM walking [count] | 83.6 ± 9.9 | 76.4 ± 6.3 | 80 – 140 [63] |
| R-R interval [ms] | 834.1 ± 81.4 | 760 ± 92.2 | 600 – 1200 [64] |
| QRS duration [ms] | 144.3 ± 11.9 | 90.2 ± 14.2 | < 120 [65] |
| ST duration [ms] | 120.8 ± 10.7 | 120.4 ± 10.1 | 80 – 120 [66] |
| T duration [ms] | 220.9 ± 16.7 | 204.6 ± 19.2 | 100 – 250 [67] |

STD: standard deviation, BPM: beat per minute

- Custom system: wearable graphene textile electrodes and custom-designed ECG acquisition board
- Commercial system: pre-gelled Ag/AgCl "wet" electrodes and OpenBCI Cyton Board
- Clinical system: pre-gelled Ag/AgCl "wet" electrodes and clinical ECG monitoring systems

![Fig. 8. (a) ECG waveform recorded from a single-arm using spray printed graphene textiles showing the P-QRS-T complex, P-wave duration (red), QRS complex duration (orange), S-T segment’s duration (green), T-wave duration (blue). The total number of R-waves correspond to the heartrate and the time elapsed between two successive R-waves represent the heart-rate variability (HRV), and a custom-written offline thresholding-based algorithm automatically detects both parameters. (b) Snapshot from benchmarking BPM values between commercially available photoplethysmography (PPG) based pulse oximeter (BPM = 70) and by the developed single-arm ECG (BPM = 70).](image-url)
extracted from the P-QRS-T complex (Table 3). Accordingly, the mean heartrate for each of the previously introduced eight participants was recorded in different conditions such as sitting, standing, and walking; and average of the means and standard deviations (STD) for all participants were listed, along with the R-R interval (essential for HRV calculation) and durations of the QRS complex, ST segment, and T-wave (Table 3).

In addition, to benchmark the extracted values from the developed system against a reference system, gold-standard pre-gelled Ag/AgCl wet electrodes were also used in conjunction with a commercial general purpose biopotential signal acquisition unit (OpenBCI Cyton Board, USA) and the single-arm ECG recording experiment was repeated. Although the single-arm ECG data recorded using the SADE system (wearable graphene textiles and custom-made readout circuit in this work) and the reference system (Ag/AgCl electrodes and OpenBCI Cyton Board) were obtained from different participants, the standard deviation of all extracted parameters turn out to be similar, and also in alignment with clinically standard chest ECGs from literature [63-67], verifying the high accuracy of the developed system.

An off-the-shelf photoplethysmography (PPG) heartbeat tracker was used to benchmark the recordings for BPM measurements (Fig. 8b). During the measurements, no significant misreading (i.e., error of more than two counts) was observed between the recorded heartrate from the PPG and single-arm ECG. All the participants’ heartbeats in standing conditions are more than sitting and walking cases. Due to the experimental method that we followed, the participants were not briefed about the experiment procedure, and when they were asked to stand up after ~ 5 min of stationary sitting, this might induce anxiety and increase their average heartbeats.

Medical ECG interpretation includes assessment of the wave morphology, their intensity, and duration of the intervals of the ECG curve. The P-wave typically reflects atrial depolarization, and its evaluation is primarily based on validating whether the impulse conduction from the atria to the ventricles is normal or not [68]. Although P-wave was observed during the single-arm ECG, its intensity was poor. The QRS complex dominantly represents the depolarization of the left ventricle. Short or broad depolarization of the QRS complex is among the features that professional healthcare staff evaluate during medical diagnosis [69]. In our single-arm ECG measurements, we observed a slightly larger QRS complex duration than typical chest electrocardiograms. The S-T segment which corresponds to the plateau phase of the action potential is yet another critical feature, whereby both its magnitude and duration are essential in diagnosing different cardiac abnormalities, and in particular to identify the onset of acute myocardial ischemia [70]. The single-arm ECG slightly exceeds the typical S-T segment duration of chest ECG. On the other hand, the T-wave reflects the rapid repolarization of contractile cells and the change in this wave occurs in an extensive range of conditions [71]. In our study, the recorded T-waves from single-arm ECG matched with typical chest ECG waveforms.

3.4. Electrocardiogram recording as a screening tool for at-home sleep monitoring

A crucial consideration for the successful development of wearable electronics is their robust long-term performance which is fundamentally related to the sensor’s fabrication, front-end readout hardware design, and system packaging. To demonstrate the superior performance and comfortable use of the developed single-arm ECG monitoring system (SADE), we performed a 45-minute-long sleep study (Fig. 9). The system was worn by a 32-year-old male participant without any diagnosed cardiovascular diseases and a long-term ECG measurement was successfully performed while the participant was taking an afternoon nap. The noise of the single-arm ECG signal can be evaluated using RMS analysis, which indicates low fluctuations of the recorded ECG signal over time. Here it changed from 3.69 μV to 3.48, to 3.67, to 3.82, and finally to 3.57 μV recorded in the 8th, 16th, 24th, 32th, and 40th minutes, respectively. The coefficient of variation (CV) is 3.54%, showing a minimal change of RMS noise during 45-minute of continuous monitoring, concluding that SADE can be used for long-term healthcare monitoring, as supported by high-quality ECG recordings.

3.5. Figure of merit

After presenting the system’s feasibility, we compared our developed single-arm ECG diagnostic unit with commercially available gel-less wearable ECG units (Table 4). “Placement” refers to where the bio-potential sensing electrodes are attached. In this work, they are in contact solely with the left arm. In commercial units, they are attached to the chest area. For “usability,” the classification was based on how wearable the system is. We categorized socially-discreet systems integrated into daily clothing as “highly wearable.” If they have authentic and stylish packaging but are not embedded into everyday clothing, we categorized them as “moderately wearable”, and the others as “slightly wearable”. The developed system here has the lowest cost while still offering acceptable size and weight matching commercially available units. It stands out as a highly wearable and compact system, thanks to its unique single-arm electrode placement location. On the contrary, the other highly wearable units are in fact much larger (e.g., the size of a T-shirt) and rely on ECG acquisition from the chest which hinders their use a separate add-on accessory to ones’ existing clothing preference or

| System             | Price | Weight (g) | Placement | Size (mm) | Usage |
|--------------------|------|------------|-----------|-----------|-------|
| Gus Works → < $100 | 155  | Left Arm   | 70 × 54   | Highly    | Wearable |
| Garmin’s HRM-Pro  | $130 | 59         | Chest     | 10 × 0.3  | Moderately Wearable |
| Qardio             | $500 | 130        | Chest     | 193 × 0.12| Moderately Wearable |
| Vivalink           | → 7.5| Chest      | 90 × 20   | Moderately Wearable |
| LeMed ECG Strap    | $300 | NA         | Chest     | Slightly   | Wearable |
| Heartlin Fit ECG Shirt | $300 | NA         | Torso     | Highly    | Wearability |
| Hexoskin Pro Kit   | $650 | NA         | Torso     | Highly    | Wearability |

Fig. 9. Long-term monitoring of single-arm ECG using smart graphene textiles for 45 min during a sleep study.
We envision that with better packaging, the system presented here can be even lighter and more compact since each electrode only takes 30 × 30 mm with negligible height and weight of only ~ 50 mg.

4. Conclusion

Health monitoring of daily patient activities provides valuable information to medical staff, and portable systems are ideal candidates for remote health surveillance. In this study, we have developed two wearable single-arm electrocardiography (ECG) prototype systems with embedded graphene textile electrodes made by different coating methods: dip-coating and spray printing. Comparing the skin-electrode impedance values to commercial silver/silver chloride (Ag/AgCl) electrodes presented a very similar level of performance for the graphene textile electrodes. Furthermore, wearable systems were tested on 8 different participants to benchmark their performance. Signals taken asynchronously from Ag/AgCl and graphene textile electrodes were compared in terms of correlation percentage and signal-to-noise ratio (SNR) values; on both of these, textile electrodes performed toe-to-toe with Ag/AgCl electrodes. Average correlation values obtained from 8 participants reach ~ 90% in static conditions (sitting) and ~ 85% in dynamic conditions (walking), while SNR values were nearly equal. Spray printed electrodes performed better than dip-coated ones, indicating that spray printing is the preferred textile finishing technique for rGO textile fabrication. Among the non-conventional ECG electrode arrangements, single-arm ECG achieved continuous monitoring with less movement and high signal intensity. The integration of the developed technology will make the devices significantly cheaper and attractive for everyday applications ranging from cardiac health trackers to psychological stress monitoring and human-computer/human-machine interactions (HCI/HMI).

Funding

This work was supported in part by Sabanci University and The Scientific and Technological Research Council of Turkey (TUBITAK) grant number 20AG028 and 119C091. Professor Murat Kaya Yapici appreciates the support of the Turkish Academy of Sciences (TUBA) within the framework of the TUBA Outstanding Young Scientist Award Program (GEIP).

Author contributions

OO performed experiments, modified the acquisition circuit, wrote scripts for signal processing and graphical user interface, constructed the board enclosure, performed conductivity and skin-electrode impedance measurements. AG designed the initial acquisition unit. GA synthesized the electroconductive textiles and performed experiments. SG assisted in performing the measurements. AG, GA, and MKY analyzed the data. OO, AG, GA and MKY wrote the manuscript. OO, AG, SG, and MKY revised the manuscript. MKY conceived the research plan and designed the experiment. AG, GA, and MKY wrote the manuscript. OO, AG, SG, and MKY revised the manuscript. MKY conceptualized the research plan and designed the experiments, reviewed the manuscript, supervised and led the project.

CRediT authorship contribution statement

Ozberk Ozturk: Methodology, Measurements, Validation, Formal analysis, Investigation, Data curation, Software, Visualization, Writing – original draft, Writing – review & editing. Ata Golparvar: Methodology, Measurements, Validation, Formal analysis, Investigation, Data curation, Software, Visualization, Writing – original draft, Writing – review & editing. Gizem Acar: Measurements, Investigation, Data curation, Writing – original draft. Saygun Guler: Measurements, Investigation, Data curation, Writing – review & editing. Murat Kaya Yapici: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Visualization, Resources, Funding acquisition, Supervision, Project Administration, Writing – original draft, Writing – review & editing.

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.

Data availability

No data was used for the research described in the article.

Acknowledgments

This article is partially based on the M.Sc. thesis of G. Acar. The authors gratefully thank the participants involved in this study. The authors thank Osman Sahin for his help in acquiring the SEM image of graphene textile fibers.

Institutional review board statement

The study was conducted according to the guidelines of the Declaration of Helsinki and approved by the Ethics Committee of Sabanci University (FENS-2020–48).

Informed consent statement

The authors declare no conflict of interest.

References

[1] A. Franchi, L. Franchi, T. Franchi, Digital health, big data and connectivity: 5G and beyond for patient-centred care, Int. J. Digit. Health 1 (2021).
[2] P.P. Jayaraman, A.R.M. Forkan, A. Marshed, P.D. Hashbigh, Y.B. Kang, Healthcare 4.0: a review of frontiers in digital health, Wiley Interdiscip. Rev.: Data Min. Knowl. Discov. 10 (2020) 234–238, e1550.
[3] M. Al-Khafajy, T. Baker, C. Chalmers, M. Asin, H. Kolivand, M. Fahim, A. Waraich, Remote health monitoring of elderly through wearable sensors, Multimed. Tools Appl. 78 (2019) 24681–24706.
[4] M. Tavakoli, J. Carriere, A. Torabi, Robotics, smart wearable technologies, and autonomous intelligent systems for healthcare during the COVID-19 pandemic: an analysis of the state of the art and future vision, Adv. Intell. Syst. 2 (2020), 2000071.
[5] A. Caizzone, An ultra-low-noise micro-power PPG sensor, EPFL (2020).
[6] C. Kendir, M. van den Akker, B. Vox, J. Metsemakers, Cardiovascular disease patients have increased risk for comorbidity: a cross-sectional study in the Netherlands, Eur. J. Gen. Pract. 24 (2018) 45–50.
[7] K. Bayouny, M. Guber, A. Elshafeey, O. Mhaimeed, E.H. Dineen, F.A. Marvel, et al., Smart wearable devices in cardiovascular care: where we are and how to move forward, Nature Reviews, Cardiology (2021) 1–19.
[8] M. Alghatfri, J. Lindsay, A brief review: history to understand fundamentals of electrocardiography, J. Community Hosp. Intern. Med. Perspect. 2 (2012) 14383.
[9] A. Boukhayma, A. Barison, S. Haddad, A. Caizzone, Ring-embedded micro-power mm-sized optical sensor for accurate heart beat monitoring, IEEE Access 9 (2021) 127217–127225.
[10] A. Boukhayma, A. Barison, S. Haddad, A. Caizzone, Earbud-embedded micro-power mm-sized optical sensor for accurate heart beat monitoring, IEEE Sens. J. 21 (2021) 19967–19977.
[11] I. Iakazade, S.S. Martin, How useful is the smartwatch ECG? Trends Cardiovasc. Med. 30 (2020) 442–448.
[12] M.K. Yapici, T.E. Alkhidir, Intelligent medical garments with graphene-functionalized smart-cloth ECG sensors, Sensors 17 (2017) 875.
[13] N.K. Jacob, E. Balaban, R. Saunders, J.C. Batchelor, S.G. Yeates, A.J. Casson, An exploration of behind-the-ear ECG signals from a single ear using inkjet printed conformal tattoo electrodes, 2018 40th Annual International Conference of the IEEE Engineering in Medicine and Biology Society (EMBC), IEEE 2018, pp. 1283–6.
[14] S. Guler, A. Golparvar, O. Ozturk, M.K. Yapici, Ear electrocardiography (ECG) with soft graphene textiles for wearable applications, IEEE Sens. Lett. 6 (2019) 1–4.
[15] W. von Rosenberg, T. Chawimaalaene, V. Goverdovsky, N.S. Peters, C. Papavassiliou, D.P. Mandic, Hearables: feasibility of recording cardiac rhythms from head and ear locations, R. Soc. Open Sci. 4 (2017), 171214.
[16] J.G. Webster, Medical instrumentation: application and design, John Wiley & Sons, 2009.
[17] G. Acar, O. Ozturk, A.J. Golparvar, T.A. Elboshra, K. Bohringer, M.K. Yapici, Wearable and flexible textile electrodes for biopotential signal monitoring: a review, Electronics 8 (2019) 479.
L.G. Guex, B. Sacchi, K.F. Peuvot, R.L. Andersson, A.M. Pourrahimi, V. J. Phiri, P. Gane, T.C. Maloney, General overview of graphene: production, properties and application in polymer composites, Mater. Sci. Eng.: B 215 (2017) 9562–9578.

C. Napolitano, A. Medina, Spectrum of ST-T wave patterns and repolarization anormality as a possible predictive marker for acute inflammatory response in COVID-19 patients, Mil. Med. 186 (2021) e34–e38.

J.B. Miller, C. Sztajzel, Heart rate variability: a noninvasive electrocardiographic method to measure the autonomic nervous system, Swiss Med. Wkly. 134 (2004) S14–S22.

F. Gao, F. García, S.H. Wittels, S. Hendricks, S. Chong, Heart rate variability as a possible predictive marker for acute inflammatory response in COVID-19 patients, Mil. Med. 186 (2021) e34–e38.

J. Stazjel, Heart rate variability: a noninvasive electrocardiographic method to measure the autonomic nervous system, Swiss Med. Wkly. 134 (2004) S14–S22.

O. Ozturk et al. received his B.S. from Middle East Technical University of Ankara, Turkey in 2017 and his M.Sc. from Sabanci University of Istanbul, Turkey in 2020. Currently, he is a Ph.D. candidate in the electronics engineering program of Sabanci University. His research interests include wearable electronics, textile electrodes and MEMS system design and fabrication.
Ata Golparvar received the B.S. degree in electrical and electronics engineering from the Azad University of Tabriz, Iran in 2016, and M.Sc. degree in electronic engineering from Sabanci University, Istanbul, Turkey in 2019. Currently he is a Ph.D. candidate in the Microsystems and Microelectronics program of Swiss Federal Institute of Technology Lausanne (EPFL), Switzerland. His research interests include wearable electronics and their applications, textile electrodes, biomedical instrumentation and embedded systems.

Gizem Acar received her B.S. degree in Physics from the Middle East Technical University-METU, Ankara, Turkey in 2017, and her M.Sc. degree from Sabanci University, Turkey in 2020. During her studies, she worked part-time at METU Technopark and METU Physics department primarily on subjects related to digital design. Later in her Masters, she focused on wearable electronics and textile electrodes. She is currently a PhD student in Lancaster University, UK.

Saygun Guler completed his MSc-Biomedical Engineering at the University of Dundee. Later he worked in digital healthcare industry for two years in Southampton Science Park, UK, where he served as a biosignal engineer. He is currently a graduate researcher pursuing his PhD in SU-MEMS research group in Sabanci University, Istanbul, Turkey.

Murat Kaya Yapici received the B.S. and Ph.D. degrees in electrical engineering from Texas A&M University, College Station, TX, USA, in 2004 and 2009, respectively. From 2009–2010, he was a Postdoctoral Research Associate with the Solid State Electronics, Photonics and Nano-Engineering Laboratory at Texas A&M University. He is the director and principal investigator of Sabanci University Micro/Nano Devices and Systems Lab (SU-MEMS), a faculty member in the Electronics Engineering Program of Sabanci University, Istanbul, Turkey, and an affiliate faculty member in the Department of Electrical Engineering, University of Washington, Seattle, WA, USA. His research interests include MEMS/NEMS, nanotechnology, wearable and flexible electronics, point-of-care devices, microfluidics, acousto-optic devices for biological and medical applications, as well as semiconductor process technology, novel nanofabrication based on scanning probes and system-level integration of nanomaterials, MEMS, and CMOS. Dr. Yapici is a member of SPIE and URSI. He has been on the technical program committee of IEEE Sensors, IEEE BSN-BHI conferences.