Reduced Graphene Oxide-Polydimethylsiloxane Based Flexible Dry Electrodes for Electrophysiological Signal Monitoring

Suraj Baloda, Sashank Krishna Sriram, Sumitra Singh, and Navneet Gupta, *Senior Member, IEEE*

***Abstract*—Graphene-based dry electrodes have shown considerable promise in electrophysiological signal monitoring applications by providing a comfortable, irritant-free alternative to traditional wet electrodes. The proposed electrode was fabricated using a spray-coating technique by depositing reduced graphene oxide (rGO) on a polydimethylsiloxane (PDMS) substrate. The rGO/PDMS dry electrodes exhibit the capability to capture and transmit weak bio-electrical signals such as Electrocardiogram (ECGs) and Electromyogram (EMGs) without significant attenuation or distortion. Experimental results show that when compared to conventional wet Ag/AgCl electrodes, the fabricated rGO/PDMS electrodes measure higher-quality ECG signals with improved SNRs while offering similar contact quality and electrode-skin impedance despite being a dry electrode. The fabricated rGO/PDMS electrodes demonstrated excellent performance and applicability making them suitable for use in wearable long-term health monitoring devices.**

***Index Terms*—Bioelectrode, contact quality, electrocardiogram (ECGs), electrode-skin impedance, electromyogram (EMGs), electrophysiological signals, polydimethylsiloxane (PDMS), reduced graphene oxide (rGO), signal-to-noise ratio (SNR).**

I. Introduction

THE United Nations Sustainable Development Goals (SDGs) serve as a blueprint for achieving a better and more sustainable future by 2030. SDG-3, provide guidelines that ensure healthy lives and promote well-being at all ages [1]. SDG-3 emphasizes on making high quality of care accessible to all. The development of lightweight, simple-to-use biomedical devices for the safe and effective diagnosis and treatment of diseases would enable the same. Furthermore, cardiovascular diseases (CVDs) are the leading cause of death globally. In 2019, the world health organization (WHO) estimated that 32% of all global deaths stem from CVDs, and in particular, heart attacks and strokes [2].

The mortality rate stemming from CVDs could be significantly reduced if individuals' biopotentials such as the ECG are monitored and analyzed on a regular basis [3], [4]. Continuous and real-time monitoring of heart activities would play an important role in the early diagnosis and treatment of cardiovascular diseases [5], [6]. In particular, ECG recordings are widely used for the early detection and management of heart problems.

However, the prevailing practice is to administer the clinical ECG test on an annual basis at best, and only to a minority of the population. The time-consuming and rigorous nature of the clinical protocol further aggravates the issue. Clinical ECG measurement involves elaborate skin preparation and the placement of 10 low-impedance wet Ag/AgCl electrodes on the chest and limbs. Commercial Ag/AgCl electrodes are optimized with the goal being to obtain a high-quality short-term measurement for rigorous diagnosis of a wide variety of pathological conditions. This, however, leaves them unsuitable for long-term monitoring and inaccessible to many. Electrolytic gels contain irritants that may trigger allergic reactions in the skin [7], [8], [9], [10]. Gels evaporate over time as well, and the electrodes are in general designed for single-use only [11], [12], [13]. Owing to these limitations, considerable effort has been put into developing alternatives that can meet the requirements of long-term ECG monitoring [14], [15], [16]. High-performance dry electrodes which can offer high contact quality without gelling or skin preparation are sought after, and important properties such as reusability, flexibility, softness, adhesion, and biocompatibility are in demand [17], [18], [19].

Dry electrodes based on rigid metallic materials such as Ag, Au, Al, and Ti have been reported for ECG and EMG monitoring [20], [21], [22]. However, the low flexibility and softness, high electrode-to-skin impedance, and low biocompatibility of such metallic materials limit their usability in mobile, wearable health monitors. For these reasons, Carbon nanomaterials such as carbon nanotubes and graphene, when embedded in polymeric matrices, yield promising materials for bioelectrode design, owing to their superior conductivity, biocompatibility, and conformity [23], [24]. In particular, graphene based dry electrodes have received significant interest in recent years for ECG monitoring

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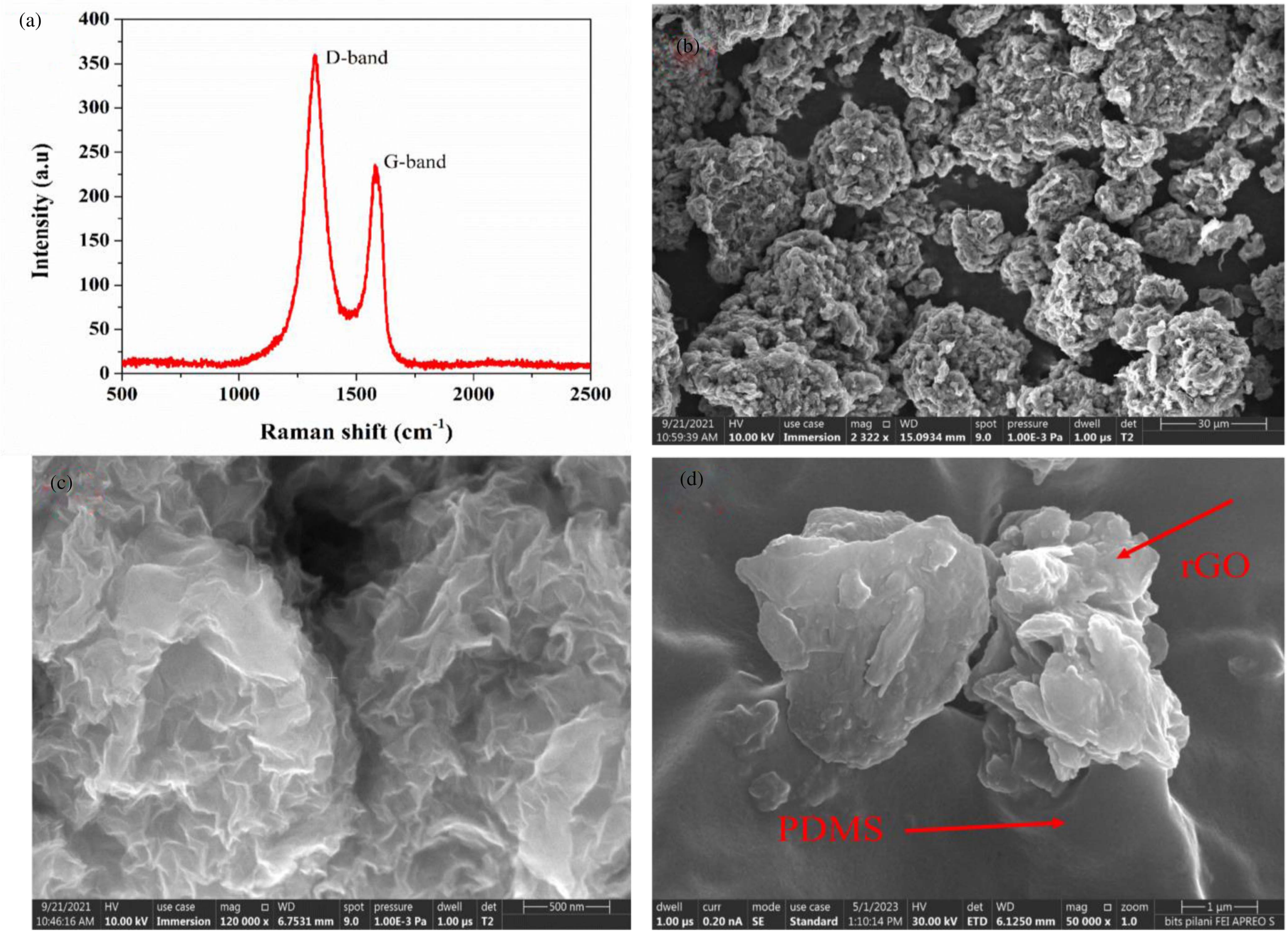

Fig. 1. (a) Obtained raman spectra of synthesized rGO, (b) FESEM image of synthesized rGO at 30 μm scale bars showing randomly distributed clusters of rGO (c) FESEM image of synthesized rGO at 500m scale bars showing nanosheets of rGO, and (d) FESEM image of rGO/PDMS dry electrode at 1μm scale bars showing rGO flakes deposited over PDMS.

applications owing to it's exceptional specific surface area and electrical conductivity, coupled with its excellent mechanical properties and thermal stability which open many doors for innovation [25], [26], [27]. However, reduced graphene oxide (rGO) has been found to exhibit superior performance owing to its enhanced sensing mechanism. This sensing mechanism operates via the oxidation of rGO and subsequent reduction of ions, a process not viable with graphene or carbon nanotubes. The difference between graphene and rGO is similar to that of the difference between metal electrodes vs Ag/AgCl.

In this study, we present a simple and low-cost flexible dry electrode for ECG and EMG monitoring, comprised of a rGO active material on a polydimethylsiloxane (PDMS) substrate. PDMS, a popular elastomeric substrate material, was chosen due to the biological and chemical inertness, flexibility, and robustness that it offers [28], [29] and the high innovation potential that it further holds in its thermal stability, high transparency, and amenability to micro-molding. The dry electrodes were fabricated using a spray coating method, where rGO was deposited onto a PDMS substrate. To assess the performance of the fabricated dry electrodes, we measured and analyzed the electrode-skin interface impedance, evaluated the quality of ECG signals measured, and benchmarked the results against commercial Ag/AgCl electrodes. The electrodes' ability to capture high-quality EMG signals is also discussed. The fabricated rGO/PDMS dry electrodes were characterized and analyzed, and the results show that the electrodes hold significant potential for application in long-term health monitoring systems, and wearable devices.

II. EXPERIMENTAL DETAILS

A. *Materials and Equipment Used*

In this work, rGO/PDMS dry electrodes were designed and fabricated. For this, rGO was synthesized by a modified hummers method and PDMS using Dow Corning's Sylgard 184 silicone elastomer kit. Materials and chemicals used in the synthesis of rGO are: Graphite fine powder (Molychem), Conc. H_2SO_4 (Molychem), Potassium permanganate $KMnO_4$ (Molychem), dilute HCl (Molychem), H_2O_2 (RANKEM), and hydrazine hydrate (RENKEM). The morphology of rGO/PDMS dry electrodes was characterized by a field emission scanning

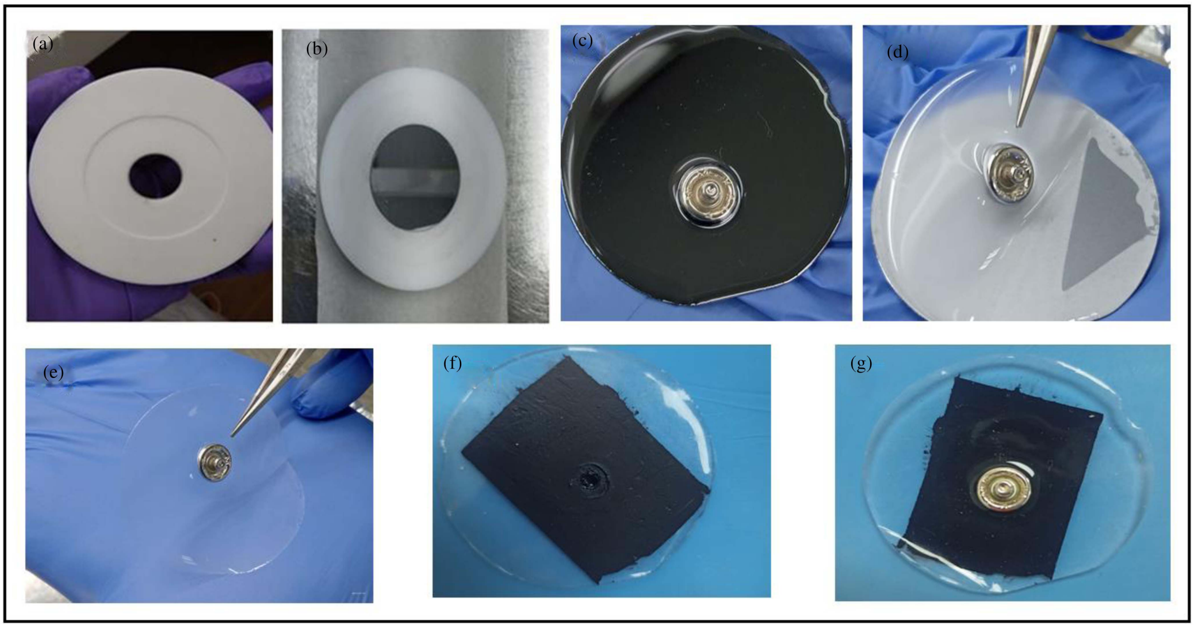

Fig. 2. The fabrication steps of the rGO/PDMS dry electrodes: (a) Teflon mold (b) silicon wafer on teflon mold (c) PDMS mixture spread on the wafer and a male snap button placed at the centre of the wafer (d), (e) peel of PDMS with male snap button (f) back-view and (g) front-view of obtained dry electrode.

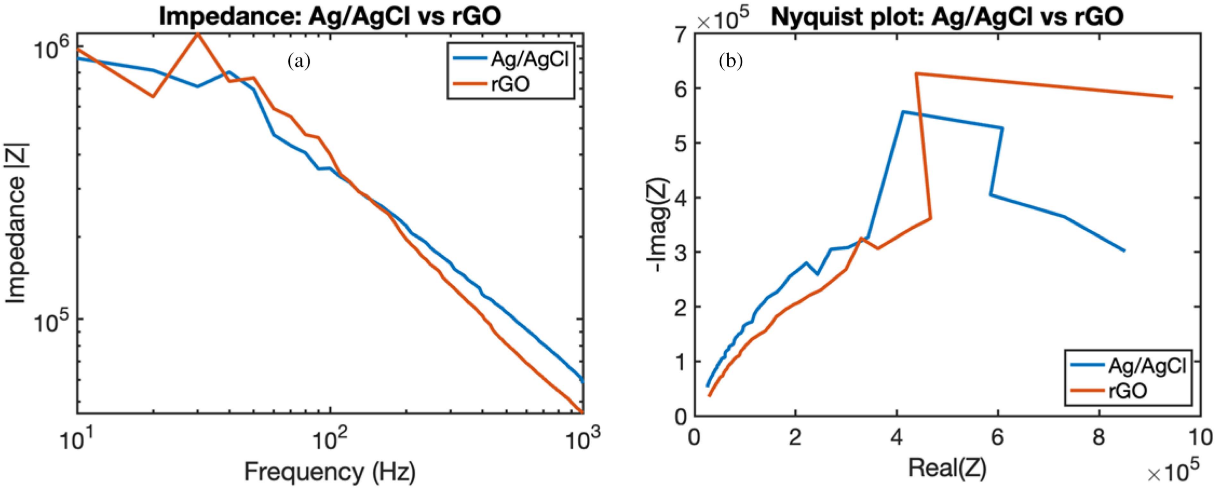

Fig. 3. (a) Measured electrode impedance for the Ag/AgCl and fabricated rGO/PDMS electrodes and its (b) nyquist plot form.

electron microscope (FESEM Quanta FEG 250). The Raman spectra were obtained with a RENISHAW InVia Raman microscope with a laser excitation wavelength of 532 nm at room temperature. Impedances were measured using a GwINSTEK 6300 LCR-Q meter. Wet Ag/AgCl ECG electrodes (EL503: 3.5 cm dia) were purchased from BIOPAC. The EKG-BTA sensor module from Vernier was used to amplify and measure the biopotentials. The ECG and EMG signals were recorded using an Arduino Uno R3 and analyzed using MATLAB.

B. Reduced Graphene Oxide (rGO) Synthesis

First, GO was synthesized from pure graphite fine powder by modified Hummer's method [30]. Sulfuric acid and phosphoric acid were mixed in a 9:1 volume ratio under continuous stirring, and 2 gm graphite powder was added to the solution. Then, 5 gm of oxidizing agent, potassium permanganate ($KMnO_4$) was added slowly, and the solution was stirred for 5 hours. To remove excess $KMnO_4$ from the solution, 10 mL hydrogen peroxide (H_2O_2) was added slowly and stirred for 20 minutes. Then, 15 mL of hydrochloric acid (HCl) and 30 mL of distilled water were added and the solution was centrifuged for 10 minutes. The excess solution was decanted away and the residual was rewashed with HCl and distilled water 5 times. Then the washed solution was dried at room temperature for 24 h to produce the GO powder. To obtain rGO, the above suspension was again sonicated for 2 h in the presence of Hydrazine hydrate and the reduction was performed at 90 °C for 1 h. The resultant solution

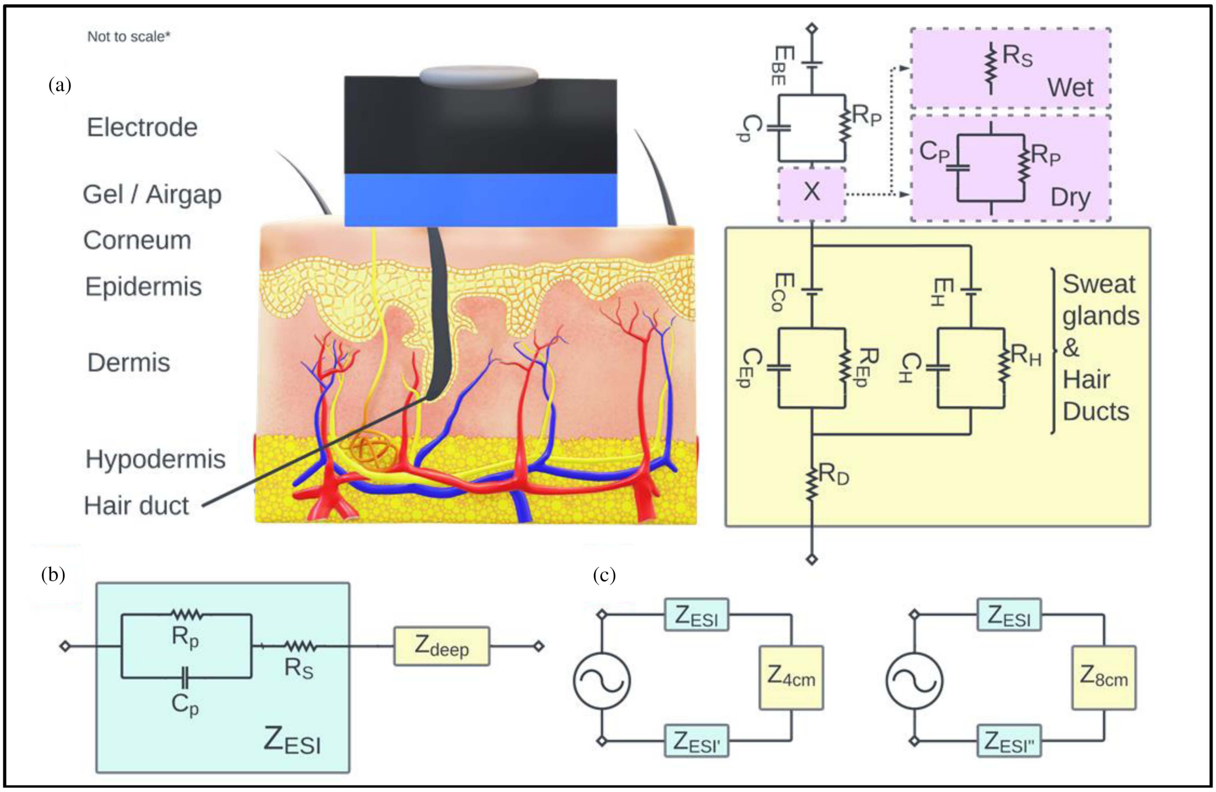

Fig. 4. (a) The electrode skin interface and its circuit model [32], [33], [34], (b) simplified circuit model [35], (c) circuit models for the measurements performed [26].

was filtered by cellulose filter paper and washed with HCl and distilled water. Finally, the filtrate was dried at room temperature for 24 h to obtain rGO.

Fig. 1(a) shows the Raman spectrum of rGO which was obtained using a 532 nm laser. Two characteristic bands, namely the D and G bands were observed at 1326 cm^{-1} and 1584 cm^{-1} respectively and are in good agreement with previous literature [31]. The presence of both D and G bands confirms the successful synthesis of rGO using the modified Hummer's method. To study the surface morphology of rGO, field-emission scanning electron microscopy (FESEM) was employed. Fig. 1(b) and (c) shows the recorded image of the synthesized rGO, showcasing randomly distributed clusters of rGO and the presence of reduced graphene oxide nanosheets. Furthermore, Fig. 1(d) illustrates the FESEM image of a fabricated rGO/PDMS dry electrode. The spray coating method was employed to deposit rGO flakes onto a flexible PDMS substrate, resulting in a dry electrode with the rGO flakes uniformly distributed over the PDMS surface.

TABLE I
EQUIVALENT CIRCUIT MODEL PARAMETERS OF THE ELECTRODE-SKIN INTERFACES FORMED

	R_P $(k\Omega)$	C_P (nF)	R_S $(k\Omega)$
Ag/AgCl	213 ± 13	1.20 ± 0.01	9.68 ± 0.44
rGO	322 ± 52	0.85 ± 0.21	28.3 ± 7.61

C. Fabrication of rGO/PDMS Dry Electrode

In this work, a simple and facile method has been employed for fabricating rGO/PDMS dry electrodes. These electrodes are composed of a PDMS substrate and an active sensing rGO layer. The fabrication process of rGO/PDMS dry electrodes is demonstrated in Fig. 2. First, a PDMS mixture was prepared by mixing the silicone elastomer base and curing agent in a 10:1 ratio. Air bubbles were removed from the mixture using a desiccator. A mold comprising a circular Teflon boat and a silicon wafer is assembled, and the PDMS mixture is poured into it. A male snap button is pressed into the mixture till it is in contact with the wafer. After allowing the mixture to settle down for 10 minutes, the setup is baked at a temperature of 90 °C for 30 minutes. The PDMS substrate is then peeled off from the silicon wafer.

Next, the rGO powder was ultrasonically dispersed in an N-methyl pyrrolidone (NMP) solution using a sonicator for one hour at 40 kHz at 100 W power. The rGO-NMP suspension formed was then cast on the backside of the PDMS substrate using a spray gun. For removing the NMP, the sample was dried using a hot air dryer. To ensure uniformity of the rGO film, the spraying and drying process is repeated several times. Thus, a rectangular $3.58 \pm 0.2 \times 2.53 \pm 0.2$ cm^2 film of rGO, of a

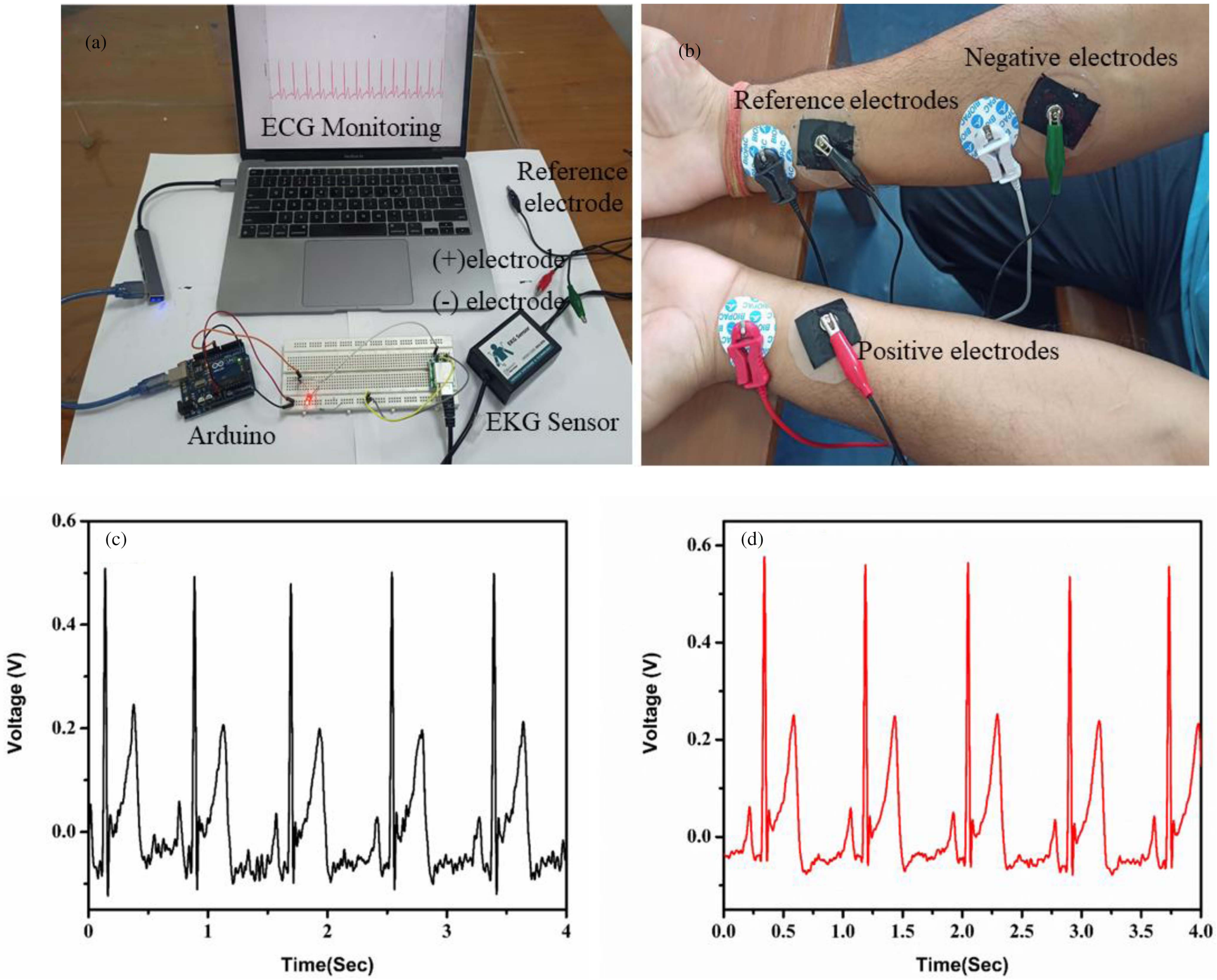

Fig. 5. (a). The electronic ECG data acquisition system and the (b) 3-electrode configuration used for the simultaneous ECG measurement using wet Ag/AgCl and dry rGO/PDMS electrodes, and ECG signals recorded using: (c) Ag/AgCl electrodes (d) fabricated rGO/PDMS dry electrodes.

size comparable to that of the Biopac Ag/AgCl electrodes, was successfully fabricated.

III. Result and Discussion

A. Impedance Measurement and Electrode Modelling

The electrode-skin interface (ESI) is critical to the performance of the electrode. Ionic currents from underneath the skin diffuse through this interface and interact with the electrodes. The exchange of electrons between the ions and electrodes are what constitute the biopotential signals we measure. To record high-quality signals with minimal distortion and noise, care must be taken to optimize the quality of the electrode contact and minimize the impedance observed [32].

In this study, we propose an approach to directly compare the impedance of the electrode-skin interfaces formed by the fabricated electrode against that of the Ag/AgCl electrode. First, electrodes were placed on the subject's forearm with spacings of 4 cm and 8 cm, and measurements were taken using a GwINSTEK 6300 LCR-Q meter over a frequency range of 10 Hz to 1 kHz. Representative samples are shown in Fig. 3.

However, the measurements also include contributions from the epidermis, dermis, deeper tissues and bodily fluids as shown in Fig. 4.

$$Z_{tot,4cm} = Z_{ESI} + Z_{ESI'} + Z_{4CM}$$

$$Z_{tot,8cm} = Z_{ESI} + Z_{ESI''} + Z_{8CM}$$

To progress, simplifying assumptions were made. Z_{ESI}, $Z_{ESI'}$ and $Z_{ESI''}$ were assumed to be identical, and the approximation $Z_{8CM} \approx 2Z_{4CM}$ was introduced. Thus, Z_{4CM} could be approximated as $Z_{tot,\,8CM} - Z_{tot,4CM}$ and could be removed from the picture. For every pair of measurements $(Z_{tot,4CM}, Z_{tot,8CM})$, we can hence estimate Z_{ESI} in 2 ways: as $Z_{tot,4CM} - Z_{4CM}$ and as $Z_{tot,8CM} - 2Z_{4CM}$. Two 4 cm and 8 cm measurements were taken for both rGO and Ag/AgCl electrodes, and 4 pairings were formed. The rGO electrodes

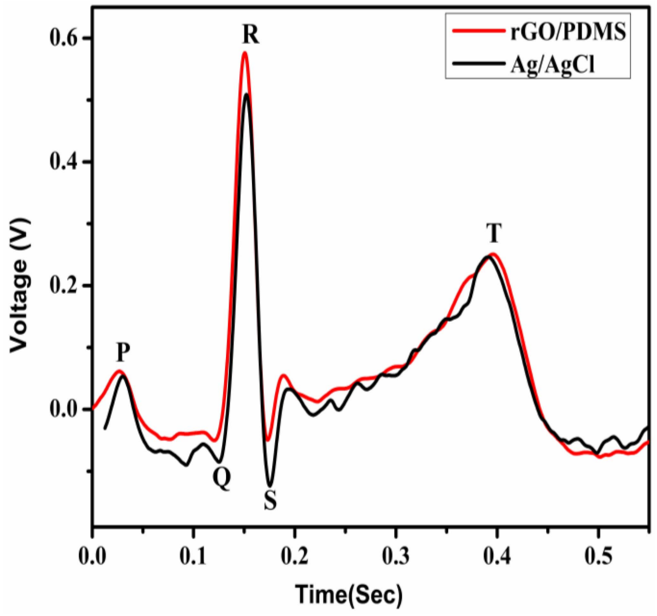

Fig. 6. Obtained ECG cycle using the rGO/PDMS dry electrodes and conventional Ag/AgCl wet electrodes.

were reused throughout the process, while Ag/AgCl electrodes were replaced after use. Hence, 8 estimates for $Z_{ESI}(f)$ were obtained. The simplified circuit model shown in Fig. 4(b) was fitted to them using the MATLAB ZfitGUI script [36], and the circuit parameters are shown in Table I.

From Fig. 3, it can be observed that the impedance offered by the rGO electrode is of the same order of magnitude as the Ag/AgCl electrode despite being a dry electrode. Table I presents a finer breakdown of the electrodes' behavior. It can be observed that on an average, R_P and $1/C_P$ of the rGO electrode are about 1.5 times higher, and the R_S about 3 times higher than that of the Ag/AgCl electrode. The absence of an electrolyte reflects strongly, especially in R_S, which models the ionic mobility across the electrode-skin interface. It must however, be noted that the high variations observed in the rGO electrodes case was also correlated with the time of measurement. Lower impedances were observed at later timestamps, perhaps due to the buildup of sweat. Considering lower bounds, the R_P, $1/C_P$ and R_S differ by factors of 1.35, 1.13 and 2.25 respectively. The impedance tradeoff involved has thus been quantified, and the potential suitability for long-term measurements is highlighted.

B. Electrocardiogram Acquisition

An ECG data acquisition pipeline was designed to sense and amplify biopotentials in general, and ECG and EMG signals in particular, as shown in Fig. 5(a). A Vernier EKG-BTA module, comprising a high-gain differential amplifier optimized for biosignals, an electrical isolation circuit, and a BTA-ELV analog protoboard adapter, was used to amplify and transmit the ECG signals.

An Arduino Uno R3 is used to sample the module's output at 1 kHz and transfer the samples to a computer over a serial interface. Sampling is done using the 10-bit ADCs offered by the ATmega328P, while UART at a baud rate of 115200 is used for data transfer. The computer runs a MATLAB script to receive and save the data from the serial interface while reinforcing the constant sampling rate of 1 kHz. Independent of the electrodes used, all measurements were performed on a healthy 28-year-old male subject at room temperature. The 3-electrode configuration shown in Fig. 5(b) is used. The subject assumed a relaxed sitting position, and no skin preparation was performed.

Fig. 5(c) and (d) show ECG waveforms measured using standard wet Ag/AgCl electrodes and the developed rGO/PDMS dry electrodes and Fig. 6. shows the zoom part of ECG waveform measured using rGO/PDMS dry electrodes and conventional Ag/AgCl wet electrodes. It was observed that both electrodes measured ECG signals with similar signal amplitudes. In the recorded signals, the features of the ECG waveform essential for diagnosing pathological conditions of the human heart, namely the P-wave, T-wave, and the QRS-complex, are clearly visible and undistorted. The features are easily extractable as well. For the recording shown in Fig. 6, the P-R interval, QRS interval, Q-T interval, and heart rate were estimated to be 0.18 s, 53.2 ms, 0.38 s, and 72.5 bpm, respectively. Owing to the clarity of the signals and the extractability of critical ECG parameters, the electrodes can also be utilized for medical diagnosis.

Furthermore, the ECG signals recorded using rGO/PDMS electrodes were observed to contain less noise than those recorded using Ag/AgCl electrodes. The signal-to-noise ratio (SNR) of the recordings was computed by first estimating the peak-to-peak amplitudes of the P-wave, T-wave, and QRS-complex (V_S), and comparing it against the peak-to-peak deviations (V_N) observed in the respective regions of the waveform [37].

$$\mathrm{SNR}_{(\mathrm{dB})} = 20\log_{10}\frac{V_S}{V_N} \tag{1}$$

The waveforms recorded using the rGO/PDMS electrodes exhibit an average SNR of 27.2 dB, while those recorded using Ag/AgCl electrodes show an SNR of 25.7 dB. Thus, the rGO/PDMS electrodes are capable of capturing less-noisy signals of similar amplitude as the Ag/AgCl electrodes, and would hence allow for improved quality of diagnosis.

C. Electromyogram Acquisition

Muscle cells generate electric potentials called electromyograms (EMG) on stimulation or activation. Analysis of EMG recordings would allow us to diagnose pathological conditions such as muscle-nerve discoordination and muscle fatigue [38]. To obtain EMG signals, two electrodes were placed across the bicep and one at the forearm without any skin preparation, and the same pipeline used to record ECG signals was utilized. EMG measurements of a 28-year-old healthy individual flexing his biceps at 3-second intervals were obtained using rGO and Ag/AgCl electrodes as shown in Fig. 7. No significant difference in quality was observed in the measurements taken using rGO and Ag/AgCl electrodes. It is to be noted that the recordings taken using the rGO/PDMS electrodes showed lower baseline variation, as can be observed in Fig. 7. Table II provides a

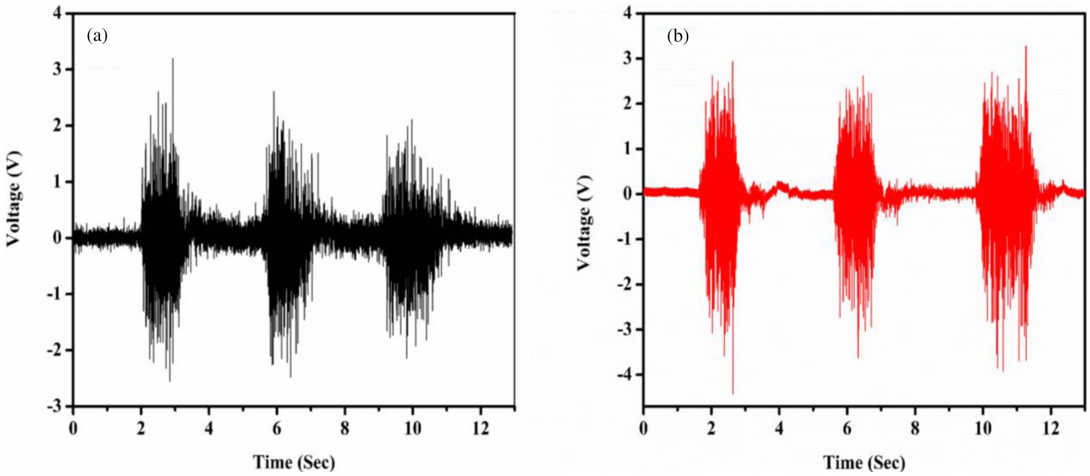

Fig. 7. EMG signals measured using (A) Ag/AgCl electrodes and (B) fabricated rGO/PDMS dry electrodes.

TABLE II
ELECTRODE MATERIALS, METHODS AND PERFORMANCE OF FLEXIBLE DRY ELECTRODES FOR ELECTROPHYSIOLOGICAL SIGNALS MONITORING

Electrode Materials	**Fabrication Method**	**Skin- Electrode Impedance range**	**SNR (dB)**	**Integration with**	**Bio signal Measured**	**Ref.**
CB/rGO/PU	Electrospinning	0.1 to 1 kHz	-----	Smart clothing	ECG and EMG	[7]
Graphene (GN)	Wet transfer and coating	20 Hz- 1 kHz	27.03	Chest-lead	ECG	[16]
Graphene sponge (GS)	Freeze-casting process	20 Hz- 1 kHz	20.3	Hand's wrist	ECG	[24]
MWCNTs/PDMS	Solution process	20 Hz- 1 kHz	24.59	Chest-lead	ECG and EMG	[39]
PDMS-CB	Solution process	0.1 to 10000 Hz	-----	Surface of left chest	ECG	[40]
Ag nanorods/RGO-PDMS	Solution process	40 Hz to 1 kHz	-----	Hand's wrist and left lower leg	ECG	[41]
Conductive Carbon	Paste	10 Hz	-----	Chest Garment	ECG	[42]
rGO coated fabric	Dyeing method	-----	21.76	Hand's wrist	ECG	[43]
rGO/PDMS	Spray-coating	20 Hz- 1 kHz	27.2	Hand's wrist	ECG and EMG	This Work

comparative analysis between the proposed dry electrodes and previous studies employing flexible dry electrodes. The findings suggest that the proposed dry electrodes demonstrate comparable electrode skin impedance and signal-to-noise ratio (SNR), affirming their suitability for detecting electrophysiological signals.

IV. CONCLUSION

In this work, we developed and evaluated rGO/PDMS dry electrodes as an alternative to Ag/AgCl electrodes for bio-signal measurements. The dry electrodes were fabricated through a simple process involving the synthesis of rGO using the modified Hummer's method and spray-coated onto a PDMS substrate. The fabricated electrodes demonstrated the ability to measure high-quality ECG and EMG signals from which clinically significant features could be extracted. When compared to Ag/AgCl electrodes, the rGO/PDMS electrodes exhibited superior performance, higher signal-to-noise ratios for ECG signals and lower baseline variations for EMG signals. On an average, the SNR was measured at 27.2 dB for the fabricated dry electrodes and 25.7 dB SNR recorded with Ag/AgCl electrodes. The electrode-skin interface impedance was analyzed using circuit fitting techniques, and the trade-off of using the rGO/PDMS electrodes was quantified. The advantages of dry electrodes in terms of convenience, comfort, durability, and ease of use make them highly suitable for long-term monitoring of ECG and EMG signals. The fabricated flexible rGO/PDMS electrodes demonstrated excellent performance and opens a pathway for preparing new epidermal devices and smart wearable systems, which can have promising applications in wearable electronics and mobile healthcare technologies.

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