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Preparation and Characterization of Silk Fibroin-PANI Nanocomposites and Their Application for Electrophysiological Signals Recording

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ABSTRACT

Wearable dry electrodes are required for long-term biopotential recordings, but their availability is limited. The diagnosis of coronary heart disease is possible with ambulatory electrocardiography (ECG). For ambulatory ECG sensing, on-skin electrodes are used by conformably contacting the skin's moving and arbitrary formed surface. However, the low skin-adhesion of electrodes restricts their use in ambulatory ECG sensing for an extended period of time. Here, it was possible to create extremely skin-adhesive and washable on-skin electrodes by developing a new composite of Poly aniline (PANI) and silk fibroin (SF). Self-adhesive property of silk fibroin - PANI electrodes was achieved by coating with silk/ Ca2+ adhesive layers. These electrodes were applied to the skin to capture high-quality ECG readings for the detection of cardiac real status in order to show how they can be used to detect precise and dependable signals. Silk fibroin - PANI electrodes showed an excellent performance for ECG signals recording over different physiological states.

INTRODUCTION

In clinical settings, ambulatory electrocardiography (ECG) is approved for the detection of coronary heart disease (CHD), the top cause of mortality for people worldwide (Poblete *et al.*, 1978) and (Poirier *et al.*, 2006). In contrast to the typical ECG's brief second recording, this method uses non-invasive electrodes to capture a long-term voltage change of the skin surface during daily activities (Steinberg *et al.*, 2019) and (Bissinger, 2017). ECG and other important vital electrophysiological signs, such as electromyograms (EMG) and electroencephalograms (EEG), are often recorded for a brief amount of time at a hospital. However, homecare has garnered a lot of interest due to the high expense of hospital-centered care and the advantages of long-period recording (Gruetzmann *et al.*, 2007) and (Chi *et al.*, 2010a), which might be realized with the advancement of wearable technology and wireless communications.

Currently, Ag/AgCl gel electrodes are the most common in a clinical context to get surface biopotentials, but they are susceptible to signal deterioration over time with continuous monitoring because the liquid in the gel electrolyte volatilizes and skin irritation occurs (Sekitani *et al.*, 2016). The creation of skin-friendly dry electrodes for biopotential measurements has thus received a lot of attention (Ferrari *et al.*, 2018) and (Hu *et al.*, 2018). Dry contact electrodes and dry capacitive (non-contact) electrodes are the two main categories for dry electrodes in literature (Chi *et al.*, 2010b). Disposable Ag/AgCl electrodes are known as wet electrodes, are frequently employed. These electrodes rely on a conductive gel to maintain excellent electrical contact with skin, but as the gel dehydrates over time, the signal quality suffers (Portelli & Nasuto, 2017a), therefore, these disadvantages limit the long-term performance of Ag/AgCl electrodes (Das *et al.*, 2020). Due to these issues, researchers are looking for substitute electrodes that may do away with the gel and even skin preparation while still producing high signal quality for an extended period of time.

However, there are now many chances to enhance the wearability of electronic devices thanks to recent developments in flexible and stretchable electronics (Zhang *et al.*, 2019), (Liu *et al.*, 2019) and (Yao *et al.*, 2019b). Stretchable on-skin electrodes for recording EP signals, such as nanocomposite EP sensors (Guo *et al.*, 2019), electronic tattoo sensors (Wang *et al.*, 2019), and epidermal electronics (Kim *et al.*, 2011a) and (Xu *et al.*, 2014), are of special interest because they permit close connection with human skin for long-term, high-fidelity monitoring. However, in order to create the previously reported on-skin EP sensors, expensive materials, advanced techniques, and specialized tools were frequently needed (Jeong *et al.*, 2013), (Kim *et al.*, 2011b), and (B. Xu *et al.*, 2015).

Due to their ability to adapt to rough and even distorted skin, flexible conductive polymer composites and inherently conductive polymers have been the focus of research on dry contact electrodes (Lee *et al.*, 2019) and (Pani *et al.*, 2018). Elastomers with conductive nanofillers such metals (Chen *et al.*, 2014), nanotubes (Lai *et al.*, 2019), nanowires (J. H. Kim *et al.*, 2018), and nanosheets (Kabiri Ameri *et al.*, 2017) make up the conductive polymer composites. Since they make up a small percentage of the elastomer matrix, conductive nanofillers have little actual interaction with human skin. Because of this, a considerable impact may be seen on the biopotential signals (S. M. Lee *et al.*, 2014). If the dry electrodes are sticky to human skin, the mismatch between a dry electrode

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and human skin can be reduced during movement. Stretchable and sticky polymer composite patches with bio-inspired micro-pillar or sucker-like structures are possible (T. Kim *et al*., 2016) and (Chun *et al*., 2018). Despite having extremely conductive and skin-compliant qualities, modern epidermal dry electrodes nevertheless have several limitations. Biocompatibility concerns may result in inflammatory reactions since they are based on artificial polymer substrates and filler ingredients (Wang *et al*., 2019b) and (Z. Xu *et al*., 2020). For these reasons, alternatives that are highly biocompatible qualities are still required.

Because of its mechanical toughness, customizable secondary structure, all-aqueous processing, and favorable biocompatibility, silk fibroin (SF), a naturally occurring protein material made by silkworms, has been recognized as being particularly appropriate for wearable electrical applications (Hwang *et al*., 2012) and (Yin *et al*., 2018). However, under ambient circumstances, reconstituted SF films tend to be stiff and brittle. Recently, Ca2+ ions were added to the SF precursor solution to create a stretchy and silky silk membrane (Ling *et al*., 2016). Self-healing is made possible by the abundance of hydrogen and coordination bonds in the SF/Ca2+ system. The uses of SF in wearable electronics are mostly restricted to its usage as a substrate, aside from its use as the active functional material, despite the material's enormous potential for use in wearable electronics (Wang *et al*., 2019c)

PANI, an electrically conductive polymer, has drawn a lot of interest in a variety of industrial and biomedical industries due to its simple manufacturing, low cost, strong electrical conductivity, biocompatibility, low toxicity, and environmental stability (Thomas & M., 2011).

Polyaniline nanocomposites, which combine the PANI matrix with either other conducting or insulating materials, are among the most promising conductive nanocomposites. These nanocomposites are widely used in the biomedical sectors of tissue regeneration and wearable electronics because to their enhanced characteristics (Oves *et al*., 2018). Owning to these favorable properties, PANI is considered as a potential candidate for wearable electrophysiological monitoring applications.

Here, silk fibroin-PANI nanocomposites had been successfully prepared by a simple and cost-effective approach. Silk fibroin protein had been extracted from natural silk cocoons followed by mixing with PANI aquas solutions to form silk fibroin-PANI nanocomposites. Silk fibroin-PANI nanocomposite electrodes had been prepared from silk fibroin-PANI solutions by a simple solvent evaporation method.

The as-prepared silk fibroin-PANI nanocomposite electrodes showed the ability to obtain high-quality ECG signals of a volunteer demonstrating their excellent performance for electrophysiological recording. Different body states such as resting, walking, and outdoor running were utilized to study the capability of silk fibroin-PANI nanocomposite electrodes to resist motion artifact and provide a highly stable ECG signal monitoring.

EXPERIMENTAL METHODS

Materials

Silk cocoons were purchased from a local supplier. Sodium carbonate (Na2CO3), Calcium chloride (CaCl2), Lithium bromide (LiBr) and formic acid were purchased from Loba chemie. Commercial Ag/AgCl electrodes were obtained from BSS Medical Supply CO., China.

Synthesis of PANI

PANI was synthesized via a chemical oxidative polymerization method (Prasutiyo *et al*., 2020). Briefly, aniline monomer was mixed with 50 mL of toluene to create a monomer solution. The initiator solution was created by combining 1.2 g of ammonium persulfate (APS) powder with 50 mL of 1M HCl solution.

The reactor glass beaker containing the monomer solution was then concurrently filled with the initiator solution drop by drop, and the mixture was stirred for 4 hours at room temperature and the color of the mixture was seen to shift from being colorless to dark green. At last, the suspension was filtered, rinsed with DI water, and dried in the drying oven at 70 oC. An aqas solution of PANI with the desired concentration was prepared by exfoliating PANI powder into formic acid solutions by a powerful ultrasonication method.

Degumming of Silk Fibers

B. Mori silkworm cocoons were first cut into little pieces, and then 2.5 g of silk cocoons were cooked for 1 h at 100 ^{0}C in an aqueous solution of Na_2CO_3 (0.02 M). The silk fibers were then squeezed to remove excess water, rinsed two times for 20 minutes each with deionized water, and allowed to air dry for the next day at room temperature.

Preparation of Silk Fibroin Solution

Silk fibroin solution was prepared following the standard protocol (Lo Celso *et al*., 2010). Briefly, degummed silk fibroin fibers were dissolved in 9.3 LiBr solution for 3h at 65 oC. In order to purify the silk fibroin/LiBr mixture solution, a dialysis process was performed by a dialysis tube (MWCO of 12 KDa) for three days at room temperature. DI water used for changed six times during the dialysis process. Centrifugation was done to get rid of the remaining impurities and in order to obtain the purified silk fibroin solution.

Preparation of Silk Fibroin-PANI Nanocomposites

An aqas solution of PANI (0.1 Wt%) was added to silk fibroin solution with a volume ratio of 1:1 and stirred for 3 h to obtain silk fibroin-PANI nanocomposite solutions. For electrode fabrication, 5 ml of silk fibroin-PANI nanocomposite solution was solvent evaporated on a petri dish (10 cm in diameter) to obtain dry silk fibroin-PANI nanocomposite films.

After that, A solution of degummed silk/ Ca_2+ in formic acid was prepared by a weight ratio of 4:6 silk to Ca_2+. Then, silk/Ca_2+ solutions were casted onto silk fibroin-PANI nanocomposite films and dried overnight to obtain adhesive silk fibroin-PANI nanocomposite electrodes.

Monitoring of ECG Signals

ECG monitoring system waw designed by interfacing AD8232 ECG Sensor with Arduino board and observe the ECG signal on a serial plotter of Arduino IDE software. A system of three electrodes was used to collect ECG signals from a human male volunteer (25 Years) where the reference electrode was a conventional Ag/AgCl electrode and two silk fibroin-PANI nanocomposite electrodes were utilized as working electrodes.

This work was approved for human experiments by Fayoum University Supreme Committee for Scientific Research Ethics (FU-SCSRE).

Characterization Techniques

SEM device (Leo Supra 55, ZEISS) was used to study the surface structure of PANI at low vacuum mood. Specimen were prepared for SEM observation by mounting freshly prepared PANI powders onto the surface of carbon tape on a measuring grid. X- ray diffraction (XRD) device (Malvern Panalytical) was used to detect and illustrate the phase purity composition and crystal structure of PANI. Specimen were prepared for XRD analysis by hand grinding PANI powders using a mortar and pestle and then mounting onto a zero back ground holder with Reflaction-Transmission Spinner stage. Uv-Vis spectroscopy was utilized to characterize PANI, silk fibroin and silk fibroin – PANI nanocomposite solutions. Freshly prepared solutions were diluted via formic acid to has a better spectrum quality. The analysis was done in a quartz cuvette containing 2.5 ml of each measured solution at a wave length of 200-1100 nm.

RESULT AND DISCUSSION

Preparation of Silk Fibroin-PANI Nanocomposite Electrodes

The whole fabrication process of silk fibroin-PANI nanocomposite electrodes were shown in Figure. 1. Breily, faint yellow colored silk fibroin solution (Figure. 2A) was extracted from silk cocoon fibers. After that, the dark green PANI solution (Figure. 2B) was added to silk fibroin solution and mixed by stirring to form silk fibroin-PANI nanocomposites. Finally, this nanocomposite solution was solvent evaporated in order to obtain silk fibroin-PANI nanocomposite electrodes (Figure 2C). Adhesive solution of silk/Ca_2+ was coated onto the surface to obtain adhesive silk fibroin-PANI nanocomposite electrodes.

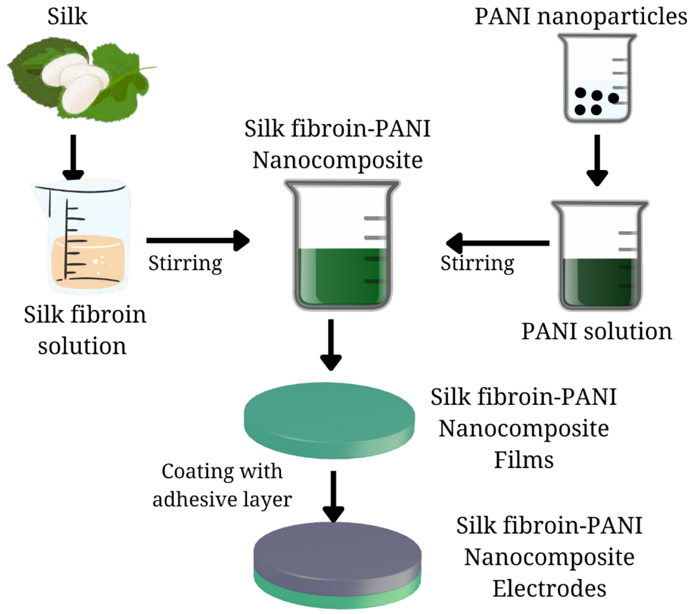


Figure 1: Silk fibroin-PANI nanocomposite electrodes preparation scheme.

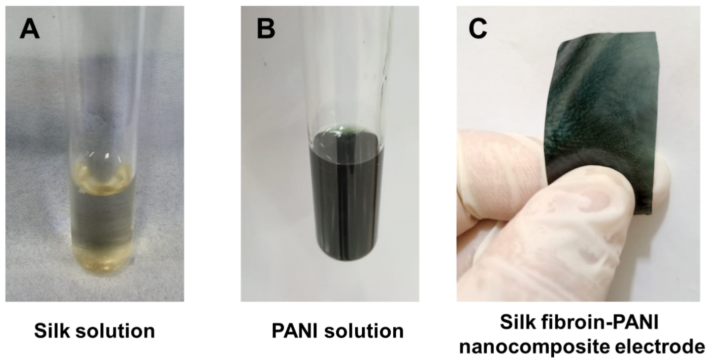


Figure 2: Digital photographs of A) Silk solution, B) PANI Solution, and C) Silk fibroin-PANI nanocomposite electrode.

Structural Characterization and Morphology of PANI

In order to study the morphology of PANI nanoparticles, field emission scanning electron microscopy images was shown in (Figure 3A-B). FESEM images of PANI nanoparticles show micro-sphere structures. In addition, flower-shaped PANI aggregates are connected together by fibril networks (Chao *et al.*, 2005). Also, EDX elemental composition (Figure 3C) of PANI nanoparticles show that PANI is composed of carbon (50.64 %) and nitrogen (49.36 %). XRD patterns of the synthesized PANI nanoparticles were shown in Figure. 4 A. The reflection lines of PANI nanoparticles appear at 2θ = 8.1°, 15.1°, 19.7°, 25.1°, and 25.9° which are corresponding to (001), (101), (100), (110), and (111) planes, respectively. In addition, Uv-vis spectroscopy results of PANI nanoparticle in DMSO solutions (Figure 4B) indicate the presence of two absorption peaks at 327 and 637 nm (Huang *et al.*, 2004). The two peaks may be related to the (π–π*) transition of the aromatic benzenoid rings and the electron excitation from the benzenoid rings of PANI, respectively (Mohammed *et al.*, 2022).

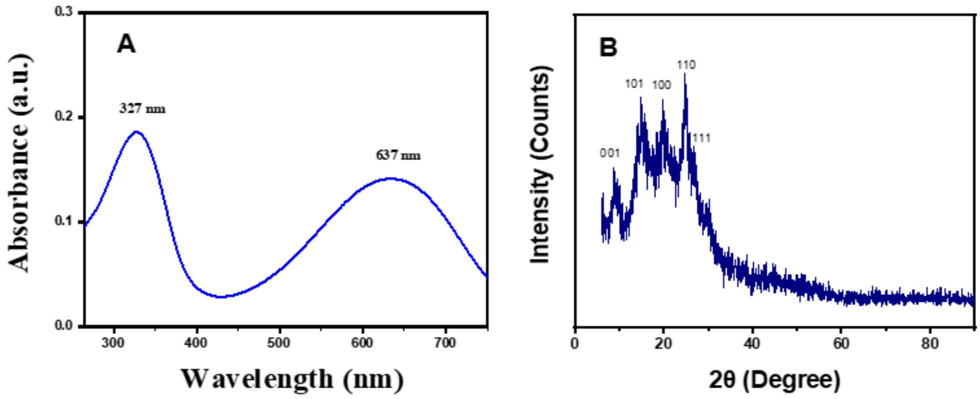
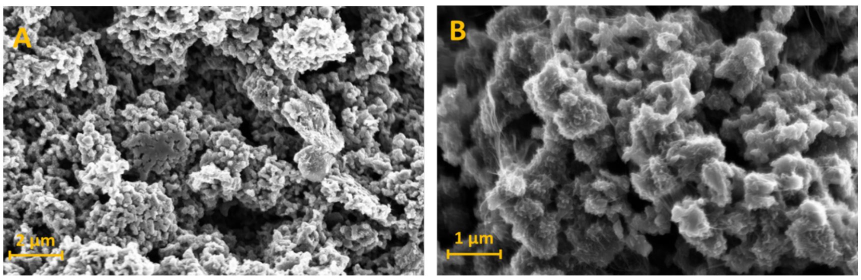


Figure 3: A) Uv-vis spectra and B) XRD patterns of PANI



C

Elements	Atomic Composition (%)
Carbon (C)	50.65
Nitrogen (N2)	49.35
Sum	100 %

Figure 4: (A,B) FESEM images of PANI nanoparticles at low and high magnification, respectively, and C) EDX elemental composition of PANI.

Silk Fibroin-PANI Nanocomposite Formation

Silk fibroin, PANI and silk fibroin-PANI nanocomposite solutions were characterized by Uv-Vis spectroscopy. As shown in Figure. 5, silk fibroin solution shows a strong absorption peak at 273 nm which is the characteristic peak of proteins (Abdel-Fattah *et al.*, 2015). Uv-Vis spectra of PANI in formic acid solutions show three absorption bands at 344, 440, and 750 nm, respectively. These absorption peaks can be related to the polaron–p* transition in the emeraldine salt, the exciton transition of the quinoid ring, a compact coiled conformation of PANI and confirms the conducting form of PANI (Dhand

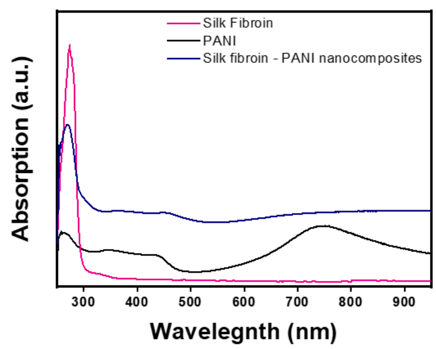


Figure 5: Uv-Vis spectra of silk fibroin, PANI and silk fibroin-PANI nanocomposite solutions.

et al., 2010). However, Uv-Vis spectra of silk fibroin – PANI nanocomposites shows the combination of both silk fibroin and also PANI absorption spectra indicating the successful formation of the nanocomposite.

Application of Silk Fibroin-PANI Nanocomposite Electrodes for Biopotential Recording

Proposed silk fibroin-PANI nanocomposite electrodes were used to assess ECG signals and compare them to traditional Ag/AgCl electrodes. The device captured every ECG signal from a a human participant male volunteer (25 Years). On the skin, the self-adhesive electrodes were positioned on chest of the volunteer.

ECG Recording at Resting Body State

The data collected utilizing traditional Ag/AgCl and proposed electrodes at a resting body state are shown in Figure. 6. The proposed silk fibroin-PANI nanocomposite electrodes were capable of collecting ECG signals effectively and the results were comparable to traditional electrodes. Moreover, by comparing the ECG signals using both electrodes, the P wave, QRS complex, and T wave, which are typical ECG wave characteristics, could be easily determined for all electrodes.

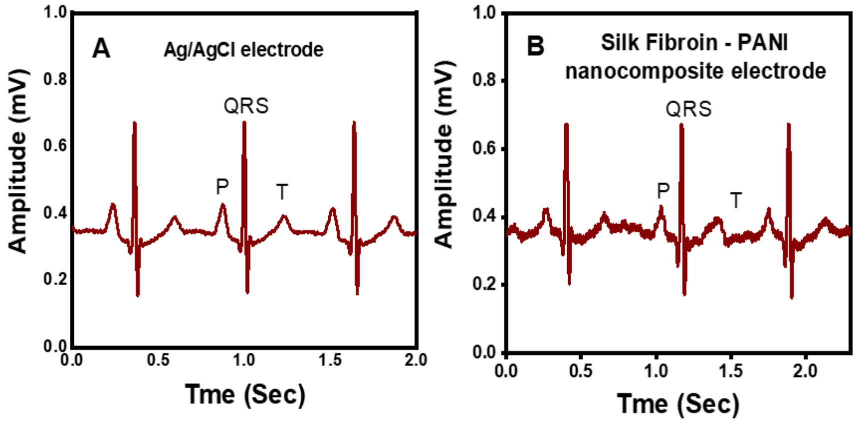


Figure 6: ECG signals collected at resting state. (A) Ag/AgCl electrodes, and (B) silk fibroin-PANI nanocomposite electrodes.

ECG Recording at Body Movement (Walking)

ECG data were collected utilizing traditional Ag/AgCl and proposed electrodes at a body movement state while walking. As shown in Figure. 7, the proposed silk fibroin-PANI nanocomposite electrodes were also capable of collecting ECG signals at walking effectively compared to traditional electrodes.

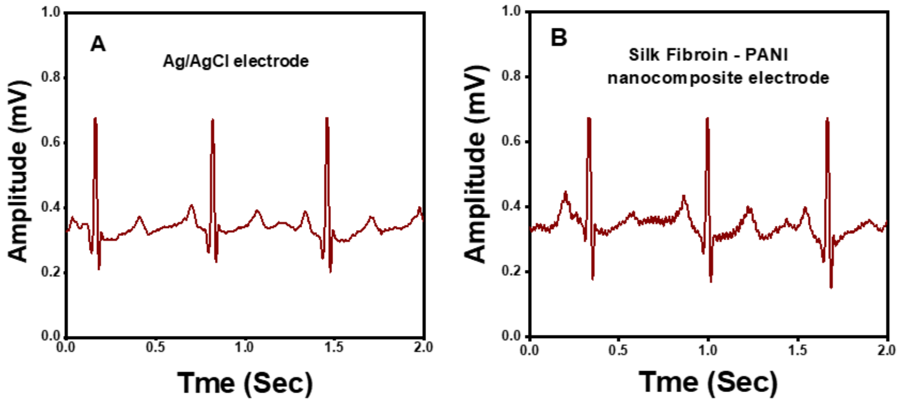


Figure 7: ECG signals collected at walking. (A) Ag/AgCl electrodes, and (B) silk fibroin-PANI nanocomposite electrodes.

ECG Recording at Body Movement (Outdoor Running)

In addition, ECG data were obtained by both, traditional Ag/AgCl and proposed electrodes at a body movement state while outdoor running. At running state, motion artifacts largely affect the quality of ECG signals, therefore a high skin conformity is required to overcome these artifacts (Peng *et al.*, 2016). ECG signals collected by traditional electrodes were deteriorated with deformations in the ECG waves due to motion artifacts (Figure. 8 A). due to gel dehydration and evaporation over time (Portelli & Nasuto, 2017b). On the other hand, the proposed silk fibroin-PANI nanocomposite electrodes were capable of collecting and maintaining a stable ECG signals while outdoor running effectively (Figure. 8 B) due to the excellent skin conformity. Therefore, silk fibroin-PANI nanocomposite electrodes can be suitable for long-term ECG signals recording.

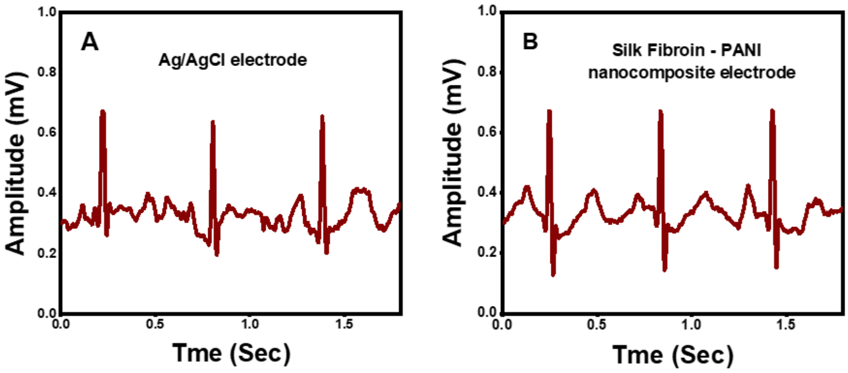


Figure 8: ECG signals collected at outdoor running. (A) Ag/AgCl electrodes, and (B) silk fibroin-PANI nanocomposite electrodes.

CONCLUSION

In this work, silk fibroin-PANI nanocomposites had been successfully fabricated using a facile and cost-effective method. Natural silk cocoons were used to extract the protein known as silk fibroin, which was then combined with PANI aquas solutions to create silk fibroin-PANI nanocomposites. A simple solvent evaporation technique had been used to create silk fibroin-PANI nanocomposite electrodes from silk fibroin-PANI solutions. The silk fibroin-PANI nanocomposite electrodes demonstrated high performance for electrophysiological recording by capturing high-quality ECG signals from a volunteer. To investigate the ability of silk fibroin-PANI nanocomposite electrodes to resist motion artefact and give a highly stable ECG signal monitoring, various body states including resting, walking, and outdoor running were used. The proposed silk fibroin-PANI nanocomposite electrodes were capable of collecting and maintaining a stable ECG signals while outdoor running effectively than traditional Ag/AgCl electrodes. Therefore, adhesive silk fibroin-PANI nanocomposite electrodes can be suitable for long-term ECG signals recording. Although our developed silk fibroin-PANI nanocomposite electrodes show a high potential for resting, walking and running states, future work may be done at a clinical point of view by studying the ability of the developed electrodes to detect several heart diseases.

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