Graphene electrodes for long-term impedance pneumography - a feasibility study

Marcel Młyńczak[1], Marek Żyliński[1], Daniel Janczak[1], Małgorzata Jakubowska[1], Wiktor Niewiadomski[2] and Gerard Cybulski[1,2]

[1] Warsaw University of Technology, Faculty of Mechatronics, Institute of Metrology and Biomedical Engineering, Warsaw, Poland
[2] Polish Academy of Sciences, Mossakowski Medical Research Center, Department of Applied Physiology, Warsaw, Poland

Abstract— **Typical disposable Ag/AgCl electrodes have potential drawbacks from a long-term measurement perspective. Particularly, they can cause an allergic reaction of the skin and can detach in case of sweating during physical activity or sleep. As they are disposable, the electrodes cannot be removed for cleaning. They must be connected to a device with separate cables, which adds to the wearer's discomfort and increases the possibility of motion artifacts. Therefore, graphene electrodes of different shapes and sizes were prepared to assess their electrode-skin contact properties compared to standard electrodes, and to evaluate the quality of obtained impedance pneumography signals and calculated tidal volumes. The results showed higher electrode-skin impedance, which did not prevent the registration of respiratory activity, after several minutes of stabilization. The accuracies of tidal volumes were comparable to the those of standard Ag/AgCl electrodes. This highlights the potential use of graphene-based, clothing-printed electrodes in cardiorespiratory and physiological healthcare, athletics, or even sleep applications.**

Keywords— **Graphene electrodes, Impedance pneumography, Long-term monitoring, Physiological measurements**

I. INTRODUCTION

There is a growing number of Holter-based applications in which electrodes are used in order to record physiology-related signals. Diagnostic electrodes can be divided into reusable and disposable ones. The first group consists of electrodes made of silver, tin, gold, sintered silver/silver chloride (Ag/AgCl), platinum or stainless steel. The second group are mainly Ag/AgCl electrodes, delivered together with adhesive gel and dry polymer foam electrodes [1, 2].

Both cardiac and respiratory activity can be recorded using standard disposable *ECG* electrodes, with Ag/AgCl layers, using impedance cardiography and impedance pneumography (*IP*) [3]. These electrodes are intended to be used for 24-h measurements, but perform best in the first few hours. They are equipped with glue, which allows for easy attachment to the body, and adhesive gel, which reduces motion artifacts and provides a very low electrode-skin contact impedance. Adhesive gel reduces the capacitive component and limits the impact of network interference on the recorded signals [4, 5, 6].

From the perspective of "dynamic" conditions appearing during natural functioning of subjects, these electrodes have some drawbacks, even preventing registration unattended by an operator. Even if the electrodes are properly positioned and attached and the skin is well prepared (via epilation, disinfection with alcohol, or removal of the stratum corneum), the clinging can change during long registration. In particular, sweat may cause slow degradation of the adhesive, leading to detachment of the electrode. The use of stronger glue is problematic, as electrodes should not damage the skin during removal. Furthermore, the size and adhesive area of the electrode, or the means of attachment, seems more important.

Also, the standard Ag/AgCl cannot be detached for and reliably reattached after bathing. The electrodes that replace it may have different properties, particularly at the beginning, and may be placed in slightly different positions, which may affect measurements. The electrodes can also cause patient discomfort through irritation of the skin or even allergic reaction (especially due to the silver and the adhesive, depending on the skin), and due to the need for a separate cable connected to each electrode. The cables may tangle during sleep, preventing further registration until the problem is addressed following an alarm.

It seems the best way to overcome the observed problems is to combine the electrodes or even integrate them with clothing, which protects against detachment by relying on pressure rather than adhesion and which makes the contact independent of perspiration. The electrode wires can then be integrated with the material and routed to the main device as a single cable [7].

Electrodes of this type should be durable, reusable and resistant to washing. It appears that graphene-based wearable electrodes, consisting of a thin layer of graphene paste with binding elements, are promising [8, 9].

Graphene layers are flexible, not prone to crumbling due to bending. Graphene generally has low resistance and high tolerance to changes in environmental conditions [10]. As graphene is pure carbon, the risk of allergic reaction is lower.

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Therefore, the first aim of this paper was **to investigate the impedance of the electrode-skin contact of graphene electrodes printed on foil in four different sizes** (in circular and square shapes), compared to classical *ECG* electrodes.

The second was **to determine the feasibility of using such electrodes for impedance pneumography measurement** by comparing the quality of *IP* signals and the accuracy of tidal volumes estimation obtained using the graphene electrodes with the optimal size to the reference method, pneumotachometry (*PNT*).

Materials & Methods and Results are divided by aim.

II. Materials and methods

AIM 1: Assessment of electrode-skin impedance

Electrode-skin impedance was measured using the bipolar method. Two electrodes were located on the arm, 4 cm apart, using medical plaster, after patient's skin preparation. A sinusoidal application current ($1\mu A$) was applied at three frequencies: 100, 200, and $400Hz$, unmatched for bioimpedance testing, because we used a USB-6001 multifunction I/O device (National Instruments, Austin, Texas, US) with the generation frequency of $1kSa/s$. Output voltage was measured with a sampling frequency of $2kHz$. All signals were acquired using Signal Express LE software (National Instruments).

Each electrode was tested for 20 minutes. Standard limb clip clamp reusable *ECG* electrodes were examined as a reference. Graphene electrodes were formed by applying graphene paste to film (125 um polyester foil) by screen printing, with graphene layer thickness of approx. $10-15\mu m$. The tested ones (Fig. 1) were circular, $1.4cm$, $2.3cm$ and $2.6cm$ in diameter, and square, with $2.6cm$ side length.

At the end of registration, the obtained results were fitted to a simple electrode-skin impedance model:

$$|Z| = \frac{1}{\sqrt{\left(\frac{1}{R}\right)^2 + (2\pi fC)^2}} \tag{1}$$

AIM 2: Tidal volume estimation accuracy

Next, the graphene electrodes were used to evaluate the quality of signals and the accuracy of tidal volumes estimated through tetrapolar impedance pneumography.

Measurements were performed with our prototype, the Pneumonitor 3. The electrode placement proposed by *Seppa et al.* was used [11, 12]. The receiving electrodes were placed on the midaxillary line at about 5th-rib level, and application electrodes on the same level on the inner arms. The Flow Measurement System (Medikro Oy, Kuopio, Finland) was used as a reference *PNT* device. The sampling frequency of the signals was 200Hz.

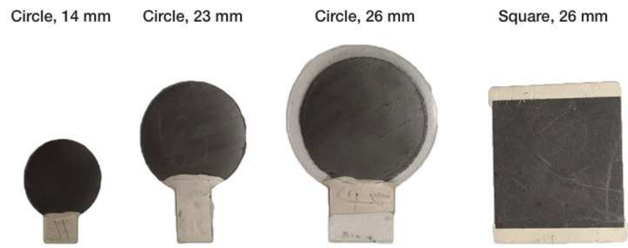

Fig. 1: Photograph of the investigated graphene electrodes.

The study participant was a generally healthy male, aged 27. We waited 5 minutes with pre-set electrodes for stabilization. Then, the subject was asked to carry out a calibration procedure consisting of 1 minute of free breathing in sitting and standing body positions. Next, he performed regular breathing at two frequencies (10 and 15 breaths per minute), with two depths (shallow and deep), in the same positions.

PNT signals were first integrated using Simpson quadrature to transform the signals into volumes. Next, both *PNT* and *IP* signals were detrended and slightly smoothed (using a moving average filter with a $0.25s$ window). We calculated two calibration coefficients (from a linear model, without considering intercept value) for the two body positions and used these for tidal volume calculations.

Each breath had two tidal volumes estimated, the first for inspiration, the second for expiration. The differences are summarized in Results.

All signal processing, analytics, and visualization were conducted using Matlab software (MathWorks, Natick, Massachusetts, US).

III. Results

AIM 1: Electrode-skin impedance assessment

We found that the values of the impedance moduli were decreasing over time and needed even 5-6 minutes to stabilize over the whole 20-minute measurements. Table 1 lists the moduli of the impedances (in $k\Omega$) measured for each electrode after 20 minutes, for all considered frequencies. The parameters of the electrode-skin impedance fitted to the results are presented in Table 2.

Table 1: The modulus of the impedance (in $k\Omega$) of each electrode, after 20 minutes of testing.

Electrode type	100 Hz	200 Hz	400 Hz
Reference ECG	11.5	6.2	3.9
Circle, 1.4*cm*	209.8	117.3	94.5
Circle, 2.3*cm*	42.8	25.3	11.6
Circle, 2.6*cm*	61.4	37.0	27.3
Square	41.1	20.5	13.1

Table 2: The parameters of the electrode-skin impedance fitted based on the test measurements.

Electrode type	A [cm^2]	R [$k\Omega$]	C [nF]
Reference ECG	8.0	122.8	71.1
Circle, 1.4*cm*	1.5	1985.4	10.2
Circle, 2.3*cm*	4.2	534.5	33.2
Circle, 2.6*cm*	5.3	597.1	31.8
Square	6.8	428.7	40.4

AIM 2: Tidal volume estimation accuracy

The post-calibration *PNT* and *IP* signals obtained from the test procedure are presented in Fig. 2 (sitting) and 3 (standing), respectively. No artifacts that would result from deterioration of electrode-skin contact, disconnection, or motion were observed. Accuracies are summarized in Table 3.

Table 3: Summary of the accuracies of tidal volumes (TVs) estimated from calibration and test signals for sitting and standing body positions.

Parameter	Sitting	Standing
R^2 of calibration model [%]	93.2	94.2
Absolute error of TV estimation [*ml*]	181.3	86.5
Relative error of TV estimation [%]	16.1	1.7
p-value of paired T-test: *PNT* vs *IP*	0.8	0.8

IV. Discussion

Dry and wet electrodes have already been tested, and the variation of their parameters over time noted [1, 2]. Our research had similar findings. The pilot studies showed that the electrode-skin impedance of graphene electrodes takes much longer to stabilize than that of silver ones. Standard reference *ECG* electrodes exhibited smaller change in contact impedance.

The size of the electrodes appears important with regard to contact impedance (undesirable) and comfort (desirable) - both increase as electrode size decreases, "on average" [13].

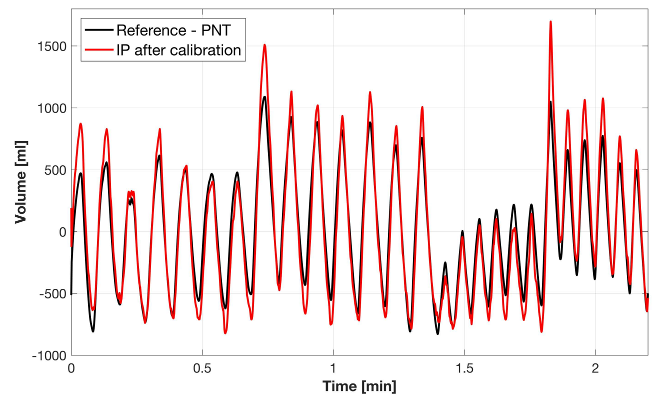

Fig. 2: The *PNT* and *IP* signals obtained through the test procedure for sitting, after calibration.

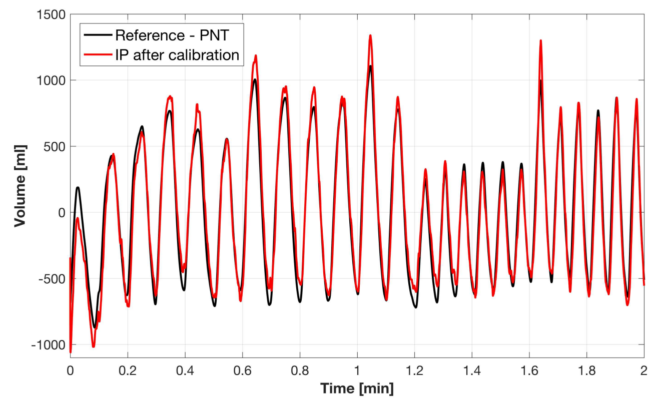

Fig. 3: The *PNT* and *IP* signals obtained through the test procedure for standing, after calibration.

Some inconsistencies could have resulted from the one-time production of the electrodes. Based on pilot results, the circular electrodes with 2.3*cm* diameter seemed to strike an optimal balance.

It seems that further enhancements of the electrode surface would not adequately reduce the impedance of the electrode-skin contact, but this should be evaluated, particularly for electrodes integrated into clothing. Similar applications have been already presented, which could allow prediction of electrode characteristics over time [14]. A properly crafted T-shirt can be worn even by the elderly. In this case, an appropriate arrangement of electrodes would not be a problem.

Interestingly, the higher electrode-skin impedance of graphene electrodes versus Ag/AgCl seemed to be a natural filter of cardiac components in impedance pneumography signals [15, 16].

A. Limitations of the study

The impedances of the electrodes were tested for a frequency range which is not utilized in classic single-frequency-based impedance pneumography and impedance cardiography. The characteristics of the impedance modulus and its change in time for higher frequencies need to be determined in the future.

Since the *IP* signals were obtained from just one subject, the analysis of accuracies is purely illustrative. However, the range of errors seemed similar to our previous analysis of a group of 10 males using standard Ag/AgCl electrodes [16]. The difference between supine and standing body positions might be caused by maintaining a more stable level of baseline impedance during standing and/or closer calibration and testing data for that body position.

The signals were acquired only in static conditions. There remains the issue of how to prepare the mounting for "dynamic" measurements while preventing motion artifacts caused by quick detachments [17].

Therefore, further plans include measurements in a larger group with electrodes integrated into a T-shirt, measurement under natural functioning conditions, and assessment of the impact of sweating on the electrical properties of the graphene electrodes.

V. CONCLUSIONS

The tested graphene electrodes exhibited greater impedance than the standard Ag/AgCl electrodes. However, it did not appear large enough to disqualify them from respiratory activity registration based on impedance pneumography.

The mean relative errors of tidal volume estimation for a single study participant were 16.1% for sitting and 1.7% for standing. They are only illustrative, but still comparable to accuracies reported in the literature.

The graphene electrodes are flexible, have flexural strength, and can be prepared with surfaces of any shape. This raises the possibility of integration with clothing and of convenient long-term cardiorespiratory analysis.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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Corresponding Author: Marcel Młyńczak
Institute: Warsaw University of Technology
Department: Faculty of Mechatronics
Address: Boboli 8, Warsaw, Poland
Email: mlynczak@mchtr.pw.edu.pl