**1. N.H. Shamsudin[1], 2. Muhammad Hidri bin Nazri[1], 3. Siti Amaniah Mohd Chachuli[2], 4. Siti Aisah Mat Junos[2], 5. MF Sulaima[1], 6. M.C. Razali[1], 7. Nursabillilah Mohd Ali[1], 8. S.S.M. Isa[3]**

Faculty of Electrical Technology and Engineering (1), Faculty of Electronic and Computer Engineering and Technology (2) Universiti Teknikal Malaysia Melaka (UTeM), Centre for Electrical Engineering Studies, Universiti Teknologi Mara, Permatang Pauh, Malaysia (3)
ORCID: 1. 0000-0002-6126-3969; 3. 0000-0001-7682-9913; 8. 0000-0003-2771-2780

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# Experimental Investigation of Graphene-Based Sensor for Exhaled Breath Performances

***Abstract***. *Exhaled breath analysis is a burgeoning research field, with a focus on the capabilities of a groundbreaking graphene-based breath sensor for disease detection. The study aims to unveil the sensor's potential as a non-invasive tool for diagnosing various health conditions by examining exhaled breath constituents. The methodology involves meticulous preparation of a graphene solution, deposition onto a silver paste electrode using a precise drop-casting method, and a critical 30-minute thermal annealing process. Characterization through Scanning Electron Microscopy (SEM) provides insights into the sensor's surface, while current-voltage (I-V) analysis explores its electrical behaviour. Discernible responses underscore the sensor's efficacy in detecting breath constituents, with impressive performance metrics, including response and recovery times, enhancing its credibility. The experimental findings affirm the commendable sensing capabilities of the graphene-based breath sensor, emphasizing its potential as a reliable tool in disease detection and contributing to the evolution of non-invasive medical technologies.*

***Streszczenie.*** *Analiza wydychanego powietrza to rozwijająca się dziedzina badań, skupiająca się na możliwościach przełomowego czujnika oddechu na bazie grafenu do wykrywania chorób. Celem badania jest odkrycie potencjału czujnika jako nieinwazyjnego narzędzia do diagnozowania różnych schorzeń poprzez badanie składników wydychanego powietrza. Metodologia obejmuje skrupulatne przygotowanie roztworu grafenu, osadzenie go na elektrodzie z pasty srebrnej przy użyciu precyzyjnej metody odlewania kroplowego oraz krytyczny 30-minutowy proces wyżarzania termicznego. Charakterystyka za pomocą skaningowej mikroskopii elektronowej (SEM) zapewnia wgląd w powierzchnię czujnika, natomiast analiza IV-V bada jego zachowanie elektryczne. Wyraźne reakcje podkreślają skuteczność czujnika w wykrywaniu składników oddechu, a imponujące wskaźniki wydajności, w tym czas reakcji i regeneracji, zwiększają jego wiarygodność. Wyniki eksperymentów potwierdzają godne pochwały możliwości wykrywania czujnika oddechu na bazie grafenu, podkreślając jego potencjał jako niezawodnego narzędzia do wykrywania chorób i przyczyniając się do ewolucji nieinwazyjnych technologii medycznych. (**Eksperymentalne badanie czujnika na bazie grafenu do pomiaru wydajności wydychanego powietrza**)*

**Keywords:** Graphene, breath sensor, Drop Casting Method, non-invasive.
**Słowa kluczowe:** Grafen, Cienka Warstwa, Metoda Osadzania Metodą Dropcast, Ludzki Oddech.

## Introduction

Analysis of exhaled breath is a significant study field that has seen a meteoric rise in interest due to recent developments in analytical methods and nanotechnology [1]-[2]. This illness detection approach does not include intrusive procedures and instead examines the exhaled breath for any traces of the disease. The study of a patient's breath may provide a non-invasive, cost-effective, real-time, qualitative, or quantitative disease diagnosis when coupled with point-of-care detection [3]. Therefore, it has the potential to replace traditional blood tests, which require entry into a living body (for example, through an incision or by inserting an instrument) and painstaking, which means the need to take pain or handle it carefully and with effort [4]. This innovation has the potential to do away with both requirements. Other volatile organic compounds (VOCs) sources include exhaled breath, sputum, and urine (all non-invasive). However, breath is the clinical sample that is easiest to handle and most straightforward to analyse [5].

Although graphene-based breath sensors have gained significant interest in health monitoring and medicinal applications, it is essential to evaluate the influence of graphene and its derivatives on human health. It is a potential 2D material that may be used in numerous applications, including developing implanted and wearable sensors for health monitoring [6]-[7]. In the detection process, a high signal-to-noise ratio may be achieved by combining factors such as high carrier mobility and density with an inherently low background noise level [8]-[9]. Since its discovery, it has garnered much interest since it has low electrical noise, a high surface-to-volume ratio, excellent chemical stability, and remarkable absorptivity [10]. Although each atom in a graphene sheet is exposed to the surrounding environment, its conductance is extraordinarily sensitive to changes in electrical and chemical circumstances [11]-[12]. This would help develop gas sensors.

Graphene-based nanomaterials have been the subject of several investigations (GB) [13-15]. However, comprehensive research on human health or environmental consequences is still lacking [2][16]. Because of present technological limitations, solid breath sensor technology remains constrained. Several constraints must be addressed before graphene is implanted into human skin, including biocompatibility, toxicity, and possible dangers. Safety evaluations are a vital component of the experiment [17]. After exposure to immune cells or biomolecules in the biological milieu, graphene's original characteristics and natural behaviors changed dramatically, perhaps leading to breakdown or biotransformation. However, the danger may be minimized by adjusting the dose dosage, form, purification, surface chemical, layers, thickness, and lateral size [4][18].

Early-stage disease diagnosis is of particular importance for effective patient identification and treatment. Exhaled human breath analysis is an up-and-coming field of research work with great potential for diagnosing diseases in non-invasive ways [19]-[20]. Breath analysis has attracted massive attention in medical diagnosis and disease monitoring. It is non-invasive, non-hazardous, and cost-effective. It reduces the time between clinical tests and the arrival of results process for disease state monitoring and environmental exposure assessment in human beings.

However, lack of patient compliance with the existing diagnostic methods limits prompt diagnosis, rendering the development of human breath [21]. The underused and marketable device is still unavailable due to multiple challenges, such as demands for a precise and repeatable experiment of targets at low concentrations. Therefore, in clinical research and various associated fields it remains

one of the top challenging tasks [3][22]. The diagnostic ability of gas-pattern detection using analytical techniques, especially sensors, is rarely used. However, it is one of the key biomarkers for several types of disease, such as breath sensors that detect exhaled volatile organic compounds and inorganic gas inside the human body [17][23].

This study exploits the use of graphene sensor at different concentration with the mixture of solvent such as Deionized Water (DI) and Ethanol (E). The fabricated sensor is exposed to various exhaled breath which are differentiate according to the nationality of the subjects which are Malay, Chinese and Indian. This experimental setup gives an early result for non-invasive nature of collecting breath samples to study an alternative approach for disease detection.

**Preparation of Graphene Sensor**

The sensing materials, graphite which at 0.01wt%, 0.02wt% and 0.05wt% concentration, was added with 10 mL of DI. The mixture was then constantly stirred for 30 minutes and undergoes sonication procedure with the ultrasonic cleaner for another 30 minutes. Similar procedure was implemented towards Ethanol.

The Kapton tape was cut with a dimension (1.4 cm x 1.9 cm) and used as the substrate. The Kapton substrates were cleansed in an ultrasonic bath of IPA solution for 10 minutes, then dried in an oven for 10 minutes. After that, the silver paste was screen printed to the Kapton substrate to make the electrode. The solution of DI and ethanol that was mixed with graphene powder has been deposited onto the Kapton substrate with a pipette to the electrode layer substrate using the drop-casting deposition method.

The samples were placed in an oven for 30 minutes to dry up the moist surface substrate, which was formed on the substrates. Finally, the dispersion on the substrate was analyzed. A good dispersion has many spots in a single location. However, a scattered dispersion was considered poor since it will not have any resistance in that area where it is scattered. Each of them would be labelled with a specific name, as shown in Table 2.

Table 2. The Variation of the Samples

| Graphene weight (wt%) | Solvent | |
|---|---|---|
| | DI | Ethanol |
| 0.01 | D1 | E1 |
| 0.02 | D2 | E2 |
| 0.05 | D5 | E5 |

**Experimental Setup of Graphene-Based Exhaled Breath Sensor**

After the analyses of the dispersion on the substrate, the gas sensor would be put inside the gas chamber. The resistivity of the sensor was checked by connecting the gas sensor inside the gas chamber to the source measure unit (SMU) model 6428 Dual Channel Picoamp meter to distinguish whether it is functional. Next, the SMU will be connected to the computer. After that, open the IV characteristic app on the computer and set the voltage from 10V to 0.1V to check the voltage breakdown.

The connection of human breath measurement is shown in Figure 2. The Tedlar bag was inflated to capture exhaled human breath. A Tedlar bag that contains human breath will connect with a 3cm hosepipe to the gas collector. Then, the LabVIEW software current measurement and the source (0.1V) was applied to the sensor inside the gas chamber and undergoes current stabilizing process for 5 minutes. Next, the air was released from the Tedlar bag into the gas chamber for one minute for a total of nine samples tested. It will flow through the gas chamber using the hosepipe (6mm) that includes the gas sensor. Last, the gas sensor response was recorded using the LabVIEW software current measurement.

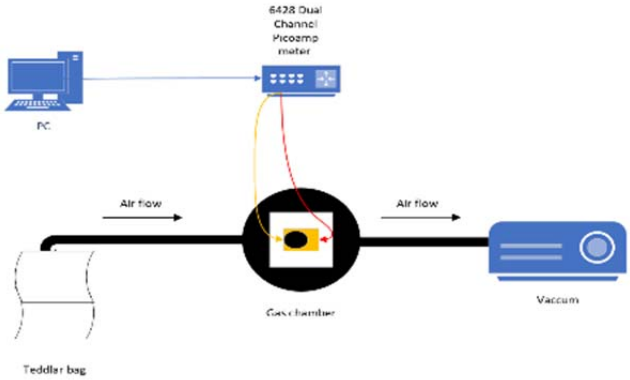



Fig.2. Connection of Human Breath Measurement

**SEM Analysis**

The structural properties of graphene solutions combined with acetone, deionized water, and ethanol is analysed using scanning electron microscopy (SEM). The shape of the produced graphene mix solution mimics flake-like particles. SEM images of the graphene mix solution indicated a well-defined exfoliated and wrinkled layered structure for the graphene as shown in Fig. 3. All these images were captured at 500 times magnification.

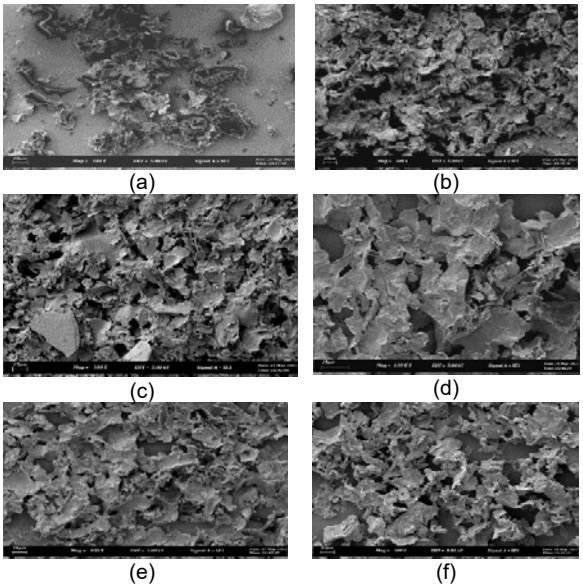



Fig.3. SEM for (a) D1, (b) D2, (c)D5, (d) E1, (e) E2 and (f) E5

**I-V Characteristic**

Current-Voltage Characteristic Curves (I-V) are a set of graphical curves used to characterise an electrical circuit's behaviour. It demonstrates the relationship between the current flowing through an electrical device and the applied voltage across its terminals. D1, D2, E1, E2 and E5 sensor has high voltage breakdown while D5 have low than 1V. So, the supply voltage for this characterization is 0.1V. Table 3 depicts the results of each sample's resistance value. Sample D5 shows the highest resistance value while E2 marked the lowest resistance value. This is attributable to both the fabrication process and the environment in which each sample is exposed.

Table 3. Resistance of sample

| Sample | Resistance (Ω) |
|---|---|
| D1 | $1.294 \times 10^3$ |
| D2 | $1.033 \times 10^3$ |
| D5 | $0.893 \times 10^9$ |
| E1 | $0.0248 \times 10^3$ |
| E2 | $7.683 \times 10^0$ |
| E5 | $0.0246 \times 10^3$ |

**Human Breath Measurement**

The sensor response, represented by S, was calculated using the equation.

$$S = Rg / Ra \quad (1)$$

where Rg and Ra are resistances with and without analyst gas, respectively. Three human breath sample tests were conducted, using female exhale breath. The samples were categorized based on nationality, as detailed in Table 4. The label of the sample goes as follows: Malay Female (MF), Chinese Female (CF) and Indian Female (IF).

Table 4. Label of the human breath sample

| Gender | Nationality | | |
|---|---|---|---|
| | Malay | Chinese | India |
| Female | MF | CF | IF |

Table 5 outlines the MF gas sensor's response characteristics. For the female breath sample, D5 in the MF breath sample shows no response which depicts in Figure 6c. Contrary to D5 sample, D1 and D2 samples exhibit a noticeable response after exposure to the breath sample.

Sensor D5's response, illustrated in Fig. 6c, indicates the current flow increases for the first 100s before stabilizing at 150s, though there is some noise. However, the sensor shows no appreciable reaction when exposed to the MF breath sample at 300s, suggesting that the D5 sensor is not responding. This lack of response could be caused by a few factors, such as insufficient interaction between the sensor surface.

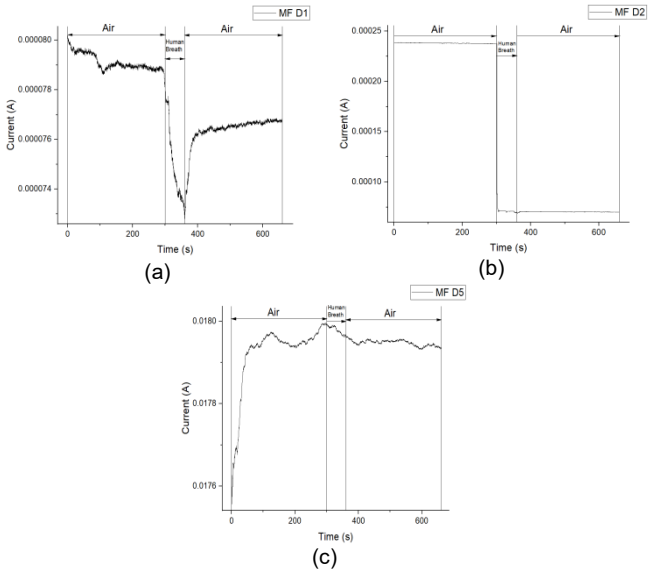



Fig.6. Human Breath Measurement of (a) MFD1, (b) MFD2 and (c) MFD5

Regarding D2's response from Fig. 6b, the initial 300s demonstrate low noise and stability. However, after exposure to the MF breath sample at 360s, it does not return to the initial resistance value. D2 exhibits a response to the breath samples within a short 4s period, highlighting a sensitivity of 0.300.

Meanwhile, D1 sample which shows in Fig. 6a has a sensitivity which is 0.930. This is recorded as an excellent sensitivity output. As the figure shows that during stability for the 300s, it has a bit of noise. After the exposure of the breath sample, it shows the result that the sensor has a response to the breath sample, but it does not recover very well. The response time takes 14s and the recovery time takes 2s only.

The performance evaluation of sensors E1, E2, and E5 was conducted based on their electrical responses to breath samples. Sample E1, as illustrated in Fig. 7a, demonstrated initial stability for a duration of 300 seconds, followed by a lack of recovery in resistance upon exposure to MF breath for 360s. In contrast, sensor D2 exhibited a rapid response of 5s to breath samples.

Meanwhile, E2 sample exhibited a remarkable sensitivity of 0.940 as depicted in the current versus time plot in Fig. 7b. This indicates a well responsive of sensor towards breath samples. Nonetheless, the sensor manifests a prolonged recovery duration, necessitating 21s to revert to its initial current baseline following exposure to a breath sample.

For sensor E5 which depicts Fig. 7c, an initial increase in current flow was observed for 100s, followed by stabilization with noise for the subsequent 150s. Notably, sensor E5 exhibited no reaction to MF breath exposure for 300s, indicating non-responsiveness. These observations provide valuable insights into the overall performance of the sensors and their potential suitability for breath sample analysis in various applications.

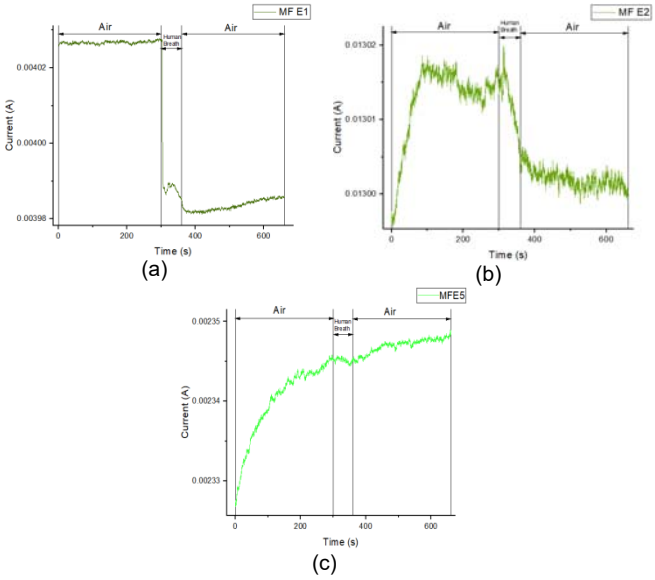



Fig.7. Human Breath Measurement of (a) MFE1, (b) MFE2 and (c) MFE5

Table 5. Characteristics of MF Sensor Response

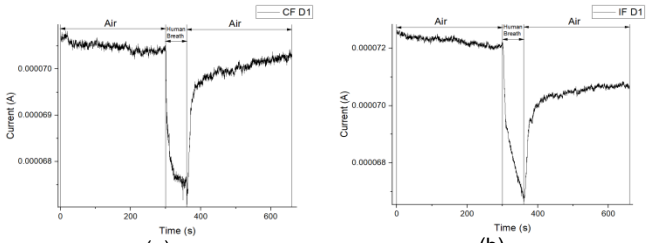
| Sample | Sensitivity | Response time | Recovery time | Recovery characteristic |
|---|---|---|---|---|
| MFD1 | 0.930 | 14s | 2s | Not recover well |
| MFD2 | 0.300 | 4s | - | Not recover |
| MFD5 | - | - | - | - |
| MFE1 | 0.990 | 5s | - | Not recover |
| MFE2 | 0.940 | 21s | - | Not recover |
| MFE5 | - | - | - | - |

The preferred sensor category, D1, was chosen for comparative analysis. CFD1 sample which is shown in Fig. 8a exhibited a sensitivity of 0.961. In the initial 300s, the sensor demonstrated lower noise and greater stability than the first D1 sensor. Upon exposure to CF breath at 300s, the sensor experienced a prolonged drop for 60s. This indicates a significant interaction between the sensor surface and the breath constituents, potentially resulting in a change in the electrical properties of the sensor material. However, it remarkably recovered to the initial value by 360s, with a response time of 30s and a recovery time of 4s.

IFD1 sample in Fig. 8b exhibited similar characteristics to CFD1 in the initial 300s, with low noise and stability. Following exposure to IF breath, the current flow drastically decreased, indicating a response to the breath sample. Although recovery was not optimal, taking 4s, it boasted the fastest response time among the D1 sensors at 7s, with a sensitivity of 0.953. This behaviour may be attributed to factors such as the kinetics of desorption or the time required for the sensor surface to revert to its original state after exposure to the breath sample.

Table 6. Characteristics of the Female Breath Sample

| Sample | Sensitivity | Response time | Recovery time | Recovery characteristic |
|---|---|---|---|---|
| CFD1 | 0.961 | 30s | 4s | Recover well |
| IFD1 | 0.953 | 7s | 4s | Not recover well |



(a) (b)

Fig 8. Human Breath Measurement of (a) CFD1 and (b) IFD1

The acquired data show different parameter performances for every sample, most likely due to differences in sensor fabrication. Variations in the concentration of graphene during deposition can result in alterations in the amount of graphene, which can then affect the active area of the sensor. Moreover, the varied reactions seen in human breath samples highlight the influence of personal factors on sensor results, including nutrition, lifestyle, and mental-emotional states. In verdict, the unique qualities of the examined participants may affect the sensor results.

**Conclusions**

In the context of human breath measurement, it is noticed that the sensor marked as CFD1 has the maximum sensitivity (0.961). The concentration of graphene in this sensor likely contributes significantly to its sensitivity, as graphene's exceptional electronic properties can enhance the sensor's response to minute variations in breath composition.

The MFD1 sensor, on the other hand, has the fastest response time, at 2s. The rapid response time of MFD1 suggests that the presence of graphene in this concentration effectively facilitates the detection and transduction of breath-related signals into measurable electrical responses.

On the other hand, although the IFD2 sensor shows the quickest recovery time of 7s, it is unable to recover to its full potential. The concentration of graphene used in the sensor's design may have an impact on this behaviour. Additionally, it might show effective kinetics of breath constituent desorption from the sensor surface.

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***Authors**: Degree student: Muhammad Hidri bin Nazri, E-mail: b012110055@student.utem.edu.my;*
*Corresponding author: Nur Hazahsha Shamsudin, Faculty of Electrical Technology and Engineering, Universiti Teknikal Malaysia Melaka (UTeM), 76100 Durian Tunggal, Melaka, Malaysia,*
*E-mail: nurhazahsha@utem.edu.my;*
*Siti Amaniah Mohd Chachuli,*
*E-mail: sitiamaniah@utem.edu.my;*
*Siti Aisah Mat Junos*
*E-mail: aisah@utem.edu.my;*
*Mohamad Fani Sulaima*
*E-mail: fani@utem.edu.my;*
*Mashitah Che Razali,*
*E-mail: mashitah@utem.edu.my;*
*Nursabillilah Mohd Ali,*
*E-mail: nursabillilah @utem.edu.my;*
*Siti Sarah Mat Isa,*
*E-mail: sitisarah6195@uitm.edu.my;*

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