

Investigation of Graphene Gas Sensor at Different Substrates for Acetone Detection

Abstract. Acetone gas is a colorless and flammable gas. Hence, it is one of the primary sources that causes combustion in high-temperature conditions. Besides, it is harmful to the health of living things. It will induce dizziness, headaches, vomiting, and irritation to the nose, eye, and throat in the short term. In the long term, it will cause damage to the central nervous system, cancer, liver, and kidney. This project aims to develop a graphene gas sensor to sense acetone and investigate the performance of the fabricated gas sensors at various thicknesses on different substrates. The substrates used are glass and Kapton film. 10 g of DI water was mixed with three different weights of graphene powder (0.01 g, 0.02 g, and 0.05 g) using a sonication bath for 30 minutes. The thickness of the sensing layer was varied through different amounts of graphene powder used in the solutions. Initially, the interdigitated electrode was deposited onto the substrates using screen-printing and annealed at 150°C for 10 minutes. After that, the sensing layer was deposited on the interdigitated electrode using the dropping technique by dropping one drop of the mixed solution and annealing at 150°C for 10 minutes. SEM and XRD characterizations are carried out to verify the sensing material of the gas sensor. The results revealed that gas sensors prepared by 0.01 g of graphene and 10 g of DI water (D-1b and D1b) produced high sensitivity to acetone compared to other samples. The gas sensor on Kapton film (D1b) had higher sensitivity than the gas sensor on the glass substrate (D-1b), with sensitivity values of approximately 7.02% and 3.24%, respectively. Sample D-2b has the shortest response time (4 s), while sample D-5b has the fastest recovery time (5 s) to acetone vapor.

Streszczenie. Aceton jest gazem bezbarwnym i palnym. Jest więc jednym z podstawowych źródeł powodujących spalanie w warunkach wysokiej temperatury. Poza tym jest szkodliwy dla zdrowia żywych organizmów. W krótkim czasie wywołuje zawroty głowy, bóle głowy, wymioty i podrażnienie nosa, oczu i gardła. W dłuższej perspektywie spowoduje uszkodzenie ośrodkowego układu nerwowego, raka, wątroby i nerek. Ten projekt ma na celu opracowanie czujnika gazu grafenowego do wykrywania acetonu i zbadanie działania wytworzonych czujników gazu o różnej grubości na różnych podłożach. Zastosowane podłoża to szkło i folia Kapton. 10 g wody DI zmieszano z trzema różnymi wagami proszku grafenowego (0,01 g, 0,02 g i 0,05 g) stosując łaźnię sonikacyjną przez 30 minut. Grubość warstwy czujnikowej była zmieniana przez różne ilości proszku grafenowego stosowanego w roztworach. Początkowo elektroda naprzemienna została osadzona na podłożach za pomocą sitodruku i wyżarzona w temperaturze 150°C przez 10 minut. Następnie warstwę czujnikową osadzano na elektrodzie naprzemienną przy użyciu techniki wkraplania przez upuszczenie jednej kropli zmieszanego roztworu i wyżarzanie w temperaturze 150°C przez 10 minut. Charakteryzacje SEM i XRD są przeprowadzane w celu weryfikacji materiału czujnika gazu. Wyniki wykazały, że czujniki gazu przygotowane z 0,01 g grafenu i 10 g wody DI (D-1b i D1b) wykazywały wysoką czułość na aceton w porównaniu z innymi próbkami. Czujnik gazu na folii kaptonowej (D1b) miał wyższą czułość niż czujnik gazu na podłożu szklanym (D-1b), przy wartościach czułości odpowiednio około 7,02% i 3,24%. Półka D-2b ma najkrótszy czas odpowiedzi (4 s), podczas gdy próbka D-5b ma najszybszy czas powrotu (5 s) do par acetonu. (Badanie czujnika gazu grafenowego na różnych podłożach w celu wykrycia acetonu)

Keywords: Graphene gas sensor, Screen-printing technique, Dropping, Kapton film, DI water

Słowa kluczowe: Czujnik gazu grafenowego, technika sitodruku, kropla, folia kaptonowa, woda DI

Introduction

Volatile organic compounds (VOC) are emitted as colorless gas from certain liquids or solids. VOC gas covers a wide range of chemical substances that are naturally occurring or man-made. Common types of VOC gasses include acetone, acetic acid, acetylene, benzene, ethanol, formic acid, methanol, isopropanol, and toluene. VOC gases come from building materials (such as carpet, paint, and composite wood products), personal care products (like cosmetics, nail removers, and hand sanitizers), and daily used equipment (like cooking gas, fuel oil, and dry cleaning). Volatile organic compounds (VOC) are flammable gases, so it is easy to cause combustion when exposed to high temperatures. ACS' Environmental Science and Technology's researchers have analyzed the level of particulate matter and VOC surrounding firefighters actively fighting fires, finding the highest exposures among hotshot teams and those establishing firebreaks [1].

Moreover, VOC gas will affect living health. Minnesota Department of Health states that exposure to high levels of VOC gas in acute term (hours to days) may cause headaches, dizziness, worsening asthma symptoms, vomiting, and irritation on the eye, nose, and throat [2]. While VOC gas exhibits a high percentage chronically (years to a lifetime), we might have symptoms like cancer, central nervous system damage, and liver and kidney damage [2]. Moreover, some VOC gas causes cancer in

animals, according to the United States Environmental Protection Agency [3].

Acetone is also categorized as one type of VOC gas. Most cosmetics and medications are made with acetone [4]. It also has a similar characteristic to VOC gases which are colorless, flammable, and can cause a negative impact on human and animal health. The literature shows that the acetone concentration in healthy people's exhaled breath ranges from 200 to 900 ppb [5]. Besides, acetone is a pollutant that enters the environment by landfill leachates, emissions from the chemical industry and other industries, automotive exhaust, and emissions [4]. Acetone also is identified as a biomarker for diabetes disease. Currently, non-invasive method for monitoring disease is widely have been used, such as gas sensor [6].

A gas sensor is needed to detect and identify this type of gas. A chemical sensor is an analyzer that selectively and irreversibly responds to a given analyte and converts input chemical quantities, such as the concentration of a single sample component or a complete composition analysis, into an electrical signal [7]. There are various gas sensors have been fabricated to detect acetone gas such as Cu_{1-x}Zn_xO [8], indium (III) sulphide [9], graphene [10], reduced graphene oxide [11], nickel oxide [12], zinc oxide [13], cerium(IV) oxide [14], agarose [15] and tin oxide [16]. Graphene has a high surface area of 2630 m²/g and ultra-high conductivity of 2.11 S/m [17]. These advantages make graphene a practical material for gas sensing in this work.

Several methods have been used to deposit the sensing layer in a gas sensor, such as spin coating [18], sputter deposition [19], drop-casting [20], screen-printing [21], spray pyrolysis [22], brush coating [16], and thermal evaporation [23]. Among them, drop-casting does not require special equipment and is low-cost for quickly sensing air contaminants at room temperature [18]. Thus, the dropping method is chosen as the deposition technique of the gas sensor sensing layer in this work.

This paper presents the fabrication of graphene gas sensors with varying thicknesses using screen-printing technology at different substrates. The graphene gas sensor was exposed to acetone vapor, and the gas sensor performance was investigated in terms of sensitivity, response time, and recovery time.

Methodology

A. Fabrication of Graphene Gas Sensor

Glass and Kapton film substrates were used as the substrate of the gas sensor. Both substrates were cut into the size of 1.5 cm x 2.0 cm. Three solutions were prepared by mixing 10 g of DI water with three different amounts of graphene powder: 0.01 g, 0.02 g, and 0.05 g. All mixed solutions were sonicated using a sonication bath for 30 minutes. The interdigitated electrode was deposited onto a substrate using a screen-printing technique and annealed at 150°C for 10 minutes in the oven. Next, one drop of the mixed solution was dropped onto the electrode using the dropping method to form a sensing layer and annealed at 150°C for 10 minutes in the oven. The flow of the gas sensor fabrication process is shown in Fig. 1. All process was applied for both substrates (glass and Kapton film).

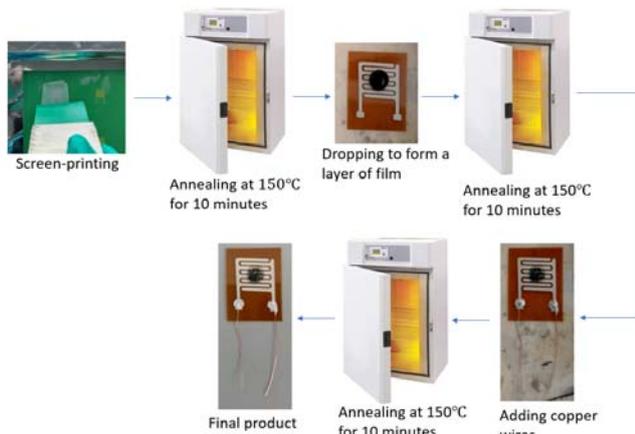


Fig.1. The flow of the gas sensor fabrication process

Table 1 lists the sample name of the graphene gas sensors based on the graphene amount and type of substrate. It can be observed that the sensing layer of the gas sensor that formed on both substrates varied slightly according to graphene amount, as displayed in Fig. 2.

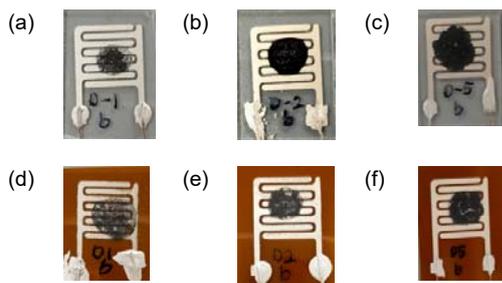


Fig.2. The fabricated graphene gas sensor (a) D-1b, (b) D-2b, (c) D-5b, (d) D1b, (e) D2b, and (f) D5b

Table 1: Sample name of the graphene gas sensors based on the type of graphene amount and substrate

	0.01 g	0.02 g	0.03 g
Glass	1b	2b	5b
Kapton film	D1b	D2b	D5b

B. Current-Voltage Measurement

Fig. 3 shows the setup of the current-voltage (IV) measurement for the gas sensor. The gas sensor was placed in a gas chamber, and supply voltage was supplied to the gas sensor using the source meter (Keithley 6487). The I-V measurement is needed to be carried out to check the conductivity of the fabricated gas sensor before exposure to the target gas. The output of the gas sensor was captured in a current form and was recorded using the LabVIEW 2010 software. From the I-V measurement, 0.1V supply voltage has been chosen as the supply voltage of the gas sensor.

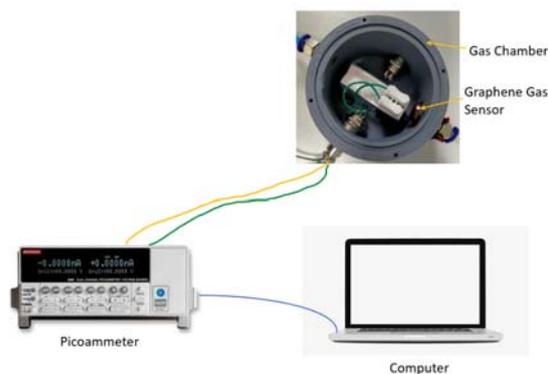


Fig.3. I-V measurement of the gas sensor

C. Experimental setup of gas sensor measurement

Fig. 4 shows the experimental setup of gas sensor measurement to acetone. Glassware is used to evaporate the acetone, and the vapor will be flowed inside the gas chamber by using a silicone hose. The acetone solution was prepared by mixing 50 ml of acetone with 50 ml of DI water using a magnetic stirrer for 5 minutes with a speed of 200 rpm. Next, the solution is heated up to 90°C for 30 minutes to create the acetone vapor.

Initially, the current of the gas sensor was observed within 5 minutes for stabilization at normal atmospheric pressure. Next, the acetone vapor was connected to the inlet of the gas chamber for 5 minutes, and the response time was observed. After 5 minutes, the hose from the glassware was disconnected from the gas chamber, and the recovery time was observed. All the data obtained from LabVIEW 2010 software were plotted using Origin 2019b software.

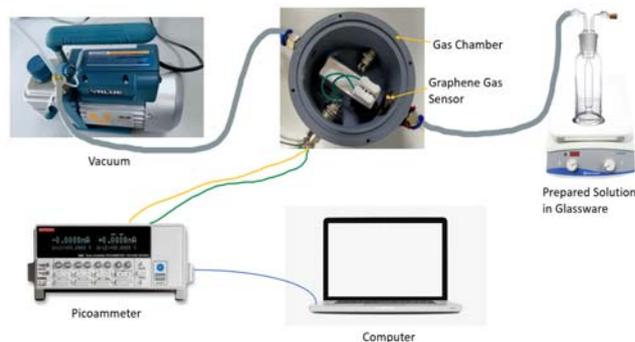


Fig.4. Experimental setup of gas sensor measurement

Result and Discussion

A. Characterization of Sensing Layer using SEM and XRD

Fig. 5 shows the morphology of the sensing layer of the gas sensor on Kapton film at magnifications of 1.0Kx. It can be observed that the structure of the graphene was flake-typed. D1b sample showed more gaps between the graphene flakes compared to the D2b and D5b samples, which can be caused by the less graphene amount dropped on the substrate. More gaps in the sensing layer allow more gas to diffuse into the sensing layer of the gas sensor; thus, a higher response will be obtained, and increased sensitivity can be produced.

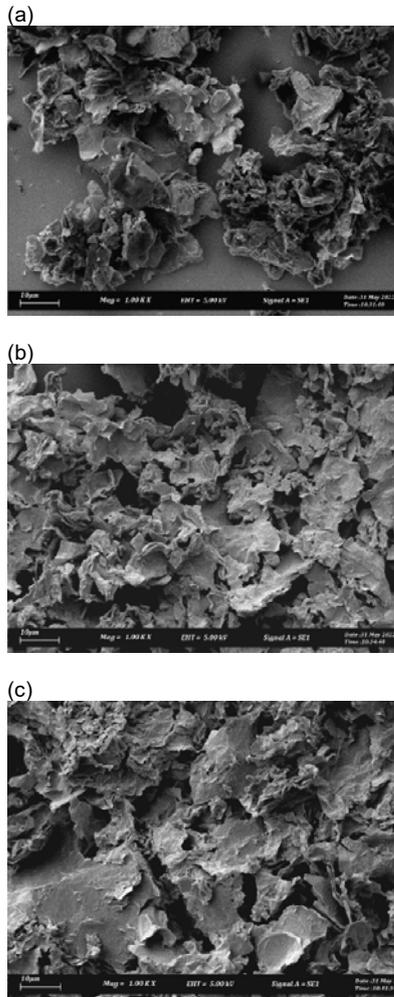


Fig.5. Morphology of the sensing layer of the gas sensor (a) D1b (b) D2b, and (c) D5b

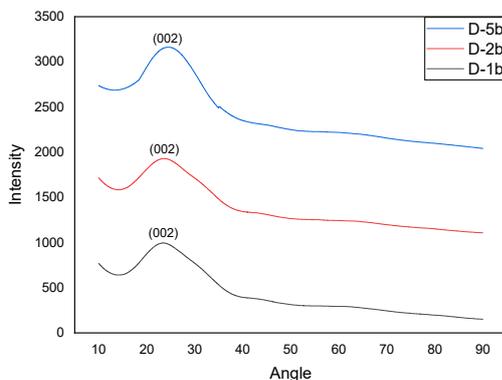


Fig.6. XRD spectra of D-1b, D-2b, and D-5b gas sensors

XRD spectra of D-1b, D-2b, and D-5b gas sensor is displayed in Fig. 6. All sensing layers detected a similar peak, where the peak was approximately at $2\theta = 23.4^\circ$, 23.6° , and 24.5° for D-1b, D-2b, and D-5b, respectively. This peak was identified as carbon, which verified that the sensing material was graphene. This result also showed that the carbon peak was broadened, thus indicating a small crystalline domain size.

B. I-V Characteristics

Fig. 7 shows I-V characteristics at 0.1V for all the fabricated gas sensors on glass and Kapton film. It can be seen that D-1b and D1b produced more linearity compared to other samples. D-5b and D5b gas sensors have the steepest slope of I-V graphs compared to the samples with 0.01 g (D1b, D-1b) and 0.02 g (D2b, D-2b) of graphene powder. This phenomenon can be attributed by the highest graphene powder (0.05g) on the sensing layer of the gas sensor. The stiffer the slope of the I-V line, the lesser the resistance value of the samples will be obtained. This is due to the slope of the IV graph being inversely proportional to the resistance. Hence, the gas sensors containing 0.05 g of graphene powder have lower resistance than others.

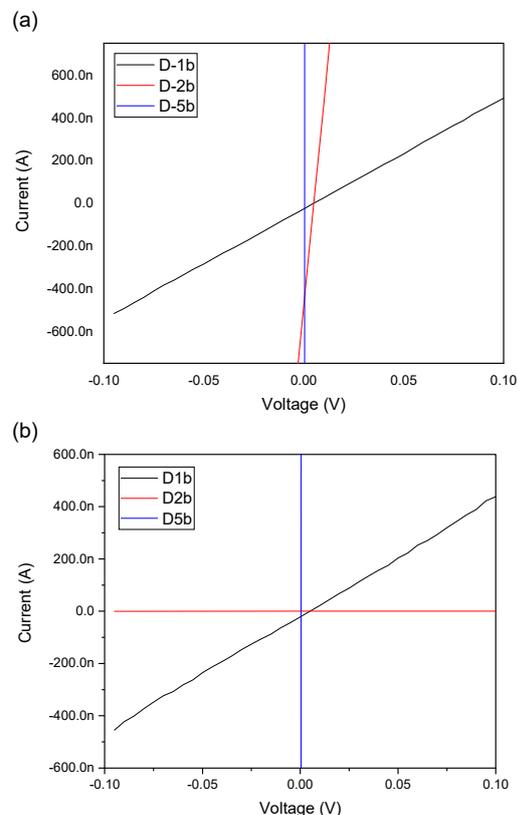


Fig.7. I-V characteristic for gas sensors using substrates (a) glass, and (b) Kapton film

Table 2 shows all the resistance of the fabricated gas sensors by applying Ohm's law, which is $R = \frac{V}{I} = \frac{1}{\text{slope of the graph}}$. It can be seen that the lowest resistance was produced by D-5b, while the highest resistance was produced by D2b. The resistance value depended on the graphene amounts placed on the sensing layer of the gas sensor. The high graphene amount contributed to the high resistance of the gas sensor. Other than that, the resistance of the gas sensor also can be influenced by the amount of silver paste amount that has been used to attach the copper wire to the leg of the electrode.

Table 2: The resistances of the gas sensors

Sample	The slope of the graph	Resistance
D-1b	5.17916×10^{-6}	193.08 k Ω
D-2b	9.52572×10^{-6}	10.50 k Ω
D-5b	0.10777	9.28 Ω
D1b	4.47976×10^{-6}	223.23 k Ω
D2b	3.10357×10^{-7}	322.21 M Ω
D5b	0.05895	16.96 Ω

C. Current Measurement

Fig. 8 and 9 show the current changes of the gas sensors when exposed to the acetone vapor. It can be observed that D-1b, D-2b, D-5b, and D1b gas sensors gave a response to acetone, while D2b and D5b gas sensors showed no reaction to the acetone. The results showed that D-1b (3.24%) and D1b (7.02%) gas sensors produced high sensitivity to the acetone. This result revealed that less graphene amount on the sensing layer helps to increase the gas adsorption for both substrates. D-2b (4 s) and D1b (5 s) gas sensors have the shortest response time compared to others. D-5b gas sensor had the fastest recovery time, approximately 5 s.

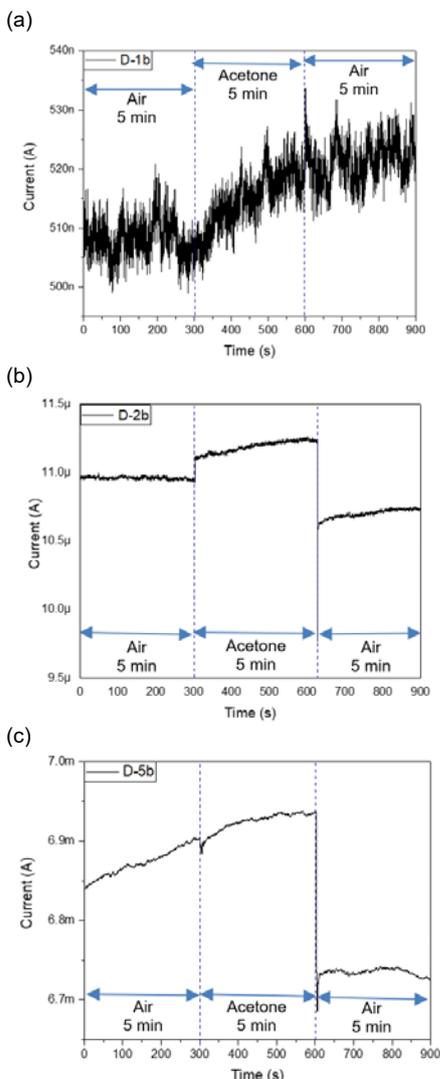


Fig.8. The current measurement of samples (a) D-1b, (b) D-2b, and (c) D-5b

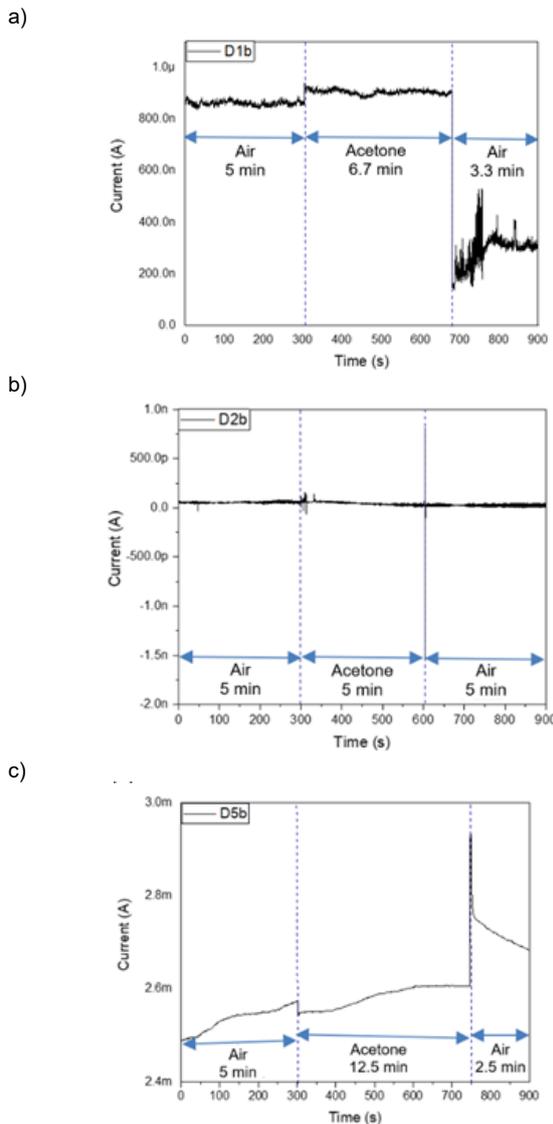


Figure 9: The current measurement of samples (a) D1b, (b) D2b, and (c) D5b

Table 3 lists the resistance value before exposure to the acetone vapor, sensitivity, response time, and recovery time of the gas sensor. The sensitivity of the gas sensor can be calculated using the equation:

$$S = \frac{R_a - R_g}{R_g} \times 100\%$$

where S represents the sensitivity of a gas sensor in percent, R_a represents the resistance of a gas sensor in air, and R_g represents the resistance of the gas sensor in the gas atmosphere [8]. The response time is obtained by finding the difference between 90% of the after-exposed gas at saturated current and 10% of the initial value at 300 s. In contrast, the recovery time is determined by finding the difference in time between the 90% after-exposed gas reading and 10% of the initial value at 600 s.

Table 3: The sensitivity, response time, and recovery time for graphene gas sensors

Sample	Resistance	Sensitivity	Response time	Recovery time
D-1b	193.08 k Ω	3.24 %	285 s	270 s
D-2b	10.50 k Ω	1.60 %	4 s	50 s
D-5b	9.28 Ω	0.47 %	230 s	5 s
D1b	223.23 k Ω	7.02 %	5 s	165 s
D2b	322.21 M Ω	-	-	-
D5b	16.96 Ω	-	-	-

Conclusion

The graphene gas sensors were successfully fabricated using screen-printing technology on glass and Kapton film. D-1b and D1b gas sensors produced higher sensitivity than other gas sensors, with sensitivity values of approximately 3.24% and 7.02%, respectively. The D-2b gas sensor had the shortest response time (4 s), while D-5b produced the fastest response time (5 s) to acetone vapor.

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