

Influence of interfacial micro-nano-modulation on the electrical properties of 2D single-layer structure

Benrui Tang^{1, #}, Yajing Wang^{2, #, *}, Jinyu Wu^{3, #}

¹ College of Physical Science and Technology, Bohai University, Jinzhou, China, 121013

² School of Physics, Henan Normal University, Xinxiang, China, 453007

³ School of Physics, Beijing Normal University, Beijing, China, 100875

* Corresponding author: 15136294111@163.com

#These authors contributed equally.

Abstract. The structure of graphene itself endows it with a variety of superior properties, and its surface functional modification has found wider applications in various fields. In this article, propose a simple surface modification method by applying a self-assembled FOTS monomolecular layer on the graphene surface via spin coating, which successfully changes the magnitude of its contact angle and thus affects the electrical properties. The electrical conductivity of the modified structures was then evaluated at different reaction times using a four-probe method. The introduction of this self-assembled monomolecular layer significantly reduced the original surface energy and lowered the average resistance for single-digit values. Combining this enhanced modified molecular layer with graphene successfully improves the electrical conductivity of this compound and the level of carrier mobility reaches the same level as that obtained by mechanical tearing. This surface engineering technique opens up new possibilities for the development of graphene-based electronic devices.

Keywords: 2D single-layer, Surface hydrophobic, High electrical conductivity, Carrier mobility.

1. Introduction

The Information Age necessitates high-speed computing and storage capabilities. Graphene's exceptional electron mobility provides it with a unique advantage, albeit at the cost of high production expenses. Graphene is a Low-dimensional carbon nanosheets composed of carbon atoms arranged in a honeycomb lattice structure with sp² hybrid orbitals. [1] Its outstanding performance stems from its singular atomic structure. Graphene exhibits remarkable electronic conductivity and boasts electron mobility that surpasses that of most similar. [2] As graphene research and fabrication methods continue to advance, its applications have expanded into diverse domains, including composite materials and electronic devices. Graphene combines several extraordinary properties, including excellent optical and electronic characteristics, a single-atom-thick two-dimensional structure, and a high specific surface area, rendering it highly appealing for a wide range of applications. [3] This paper employ a surface growth mechanism to cultivate decomposed graphene at elevated temperatures on the substrate surface. Through continuous refinement of the micro-nano surface design of graphene, then ultimately enhance the electrical properties of monolayer graphene.

Graphene, owing to its facile electronic structure modulation and exceptional conductivity, holds significant promise for application in integrated circuits [4, 5]. Nevertheless, its distinctive zero-band gap structure sets it apart from conventional semiconductors, resulting in a minimal switching ratio and limited switching properties, thereby constraining its utility in electronic devices [6]. To broaden the scope of graphene's application within the realm of integrated circuits, the manipulation and regulation of its bandgap have emerged as pivotal challenges in graphene technology. Presently, several techniques exist for bandgap modulation in graphene, including chemical doping, such as the production of large-area G-h-BN heterostructures [7], covalent bond modification, and the application of gate electric fields. The strategic adjustment of graphene's bandgap not only amplifies its switching ratio but also enables its extensive integration in the domain of integrated circuits.

The manipulation of graphene's electronic properties presents a formidable challenge, one that involves intricate adjustments to its micro/nano-structural attributes and necessitates indirect modulation of the substrate material to introduce a bandgap [8]. In light of this challenge, this paper is dedicated to the exploration of techniques for surface engineering in graphene, with a particular focus on the micro-nano structural enhancements. Chemical Vapor Deposition (CVD) is widely acknowledged for its proficiency in fabricating expansive, high-quality graphene films, primarily due to its notable controllability and scalability [9, 10]. Nonetheless, CVD-produced graphene exhibits limited electron mobility advantages compared to silicon-based materials [11]. To overcome this limitation, we have implemented chemical modification techniques in conjunction with self-assembly technology to enhance the micro-nano structure of graphene surfaces [12]. As a result, the technique succeeded in enhancing the electrical properties of graphene, and in turn changed the original band gap of graphene [13]. This endeavor capitalizes on the use of self-assembling molecules such as FDTS characterized by their low surface energy, high static contact angles, and exceptional stability [14]. The application of FDTS leads to a transformation of the surface wettability of graphene, transitioning it from hydrophilic to hydrophobic. This modification has profound implications for the micro-nano structural alterations in the graphene surface [15].

In this paper, we employed CVD to synthesize continuous monolayer graphene of exceptional quality. Subsequently, we applied FDTS, to effectuate surface modification, thereby reducing the surface energy of the graphene and rendering it hydrophobic. The alteration in graphene's surface roughness before and after this modification is evident in both the defect peak observed in the Raman spectrum and the changes in the contact angle of the graphene. Furthermore, we assessed the electrical resistance of the graphene before and after the modification using a probe platform, revealing a discernible decrease in resistance. This reduction in resistance signifies an enhancement in electron mobility within the graphene structure.

2. previous preparation

2.1. Sample preparation

The preparation method of graphene determines whether high quality graphene films can be obtained. We use CVD to prepare graphene. CVD is one of the most widely used deposition techniques for large-scale industrial preparation of thin film materials in the semiconductor industry, and the principle is to introduce two or more gaseous raw materials into a reaction chamber for chemical reaction to produce a new material is deposited on the surface of the substrate wafer.

The CVD growth of graphene involves three main aspects: carbon source, growth substrate, and growth conditions. Graphene is prepared by surface growth mechanism, using carbon compounds such as methane, through its high temperature decomposition on the surface of the substrate to grow graphene, high temperature gaseous carbon source adsorbed on the surface of the metal substrate, further nucleation and growth into "graphene islands". The "graphene islands" grow continuously to obtain a continuous graphene film.

In this experiment, methane was used as the carbon source and copper foil with high purity was used as the metal substrate. The growth of graphene started from a large number of discrete single-crystal graphene islands, and as the growth process proceeded, the graphene islands grew gradually and finally interconnected to form continuous graphene films with a very high content of monolayer graphene. The preparation of graphene by this method has the advantages of high quality of prepared graphene, good controllability and low cost.

2.2. Transfer of graphene

The graphene transfer process uses a wet transfer process to transfer graphene from copper foil to the target substrate. Since the graphene transfer process is long, the proficiency of the operation has a great impact on the quality of the subsequent graphene.

The PMMA and etching solution required for the transfer process are prepared in advance, where the whole operation of configuring PMMA is performed in a fume hood. PMMA is spin-coated on the surface of copper foil containing graphene to create mechanical support material for graphene and to prevent graphene from being torn by liquid surface tension during copper foil etching process causing breakage.

The PMMA-coated samples were baked to tightly bound the PMMA to the graphene layer and dried on a heating plate at 90°C for 5-10 min. The baked samples were immersed in 0.2 g/ml ferric chloride solution or 0.5 M of $(\text{NH}_4)_2(\text{SO}_4)_2$ solution to corrode and allowed to float on the surface of the solution for 30-60 min to completely remove Cu. After the Cu corrosion is completed the PMMA-graphene film is fished up from under the liquid surface using a PTFE spoon with a certain amount of solution kept inside the spoon to enable the film to spread on the liquid surface. Transfer the film to a Petri dish with ionized water and leave it for 3 min, wait for FeCl_3 to spread evenly in the solution, then transfer the film 4 times following the same steps, i.e., wash the film 5 times to ensure that the residual etchant is removed. The PMMA-graphene film is fished out from the deionized water using the substrate, so that the film covers the intended substrate position, and the sample is kept dry after the attachment is finished. The dried sample is baked again and heated on a heating platform at 165°C for 15 min to soften the PMMA and reduce the wrinkles formed during the transfer process, and at the same time, the graphene can be more closely attached to the substrate. Put the baked sample into a beaker with acetone and slowly heat the acetone to 70-100°C.

Repeat the above operation three times to remove PMMA, and finally put the sample into a beaker with alcohol to wash and remove the acetone, and then put the sample into deionized water to wash and remove the alcohol, and finally put the sample into an oven to dry.

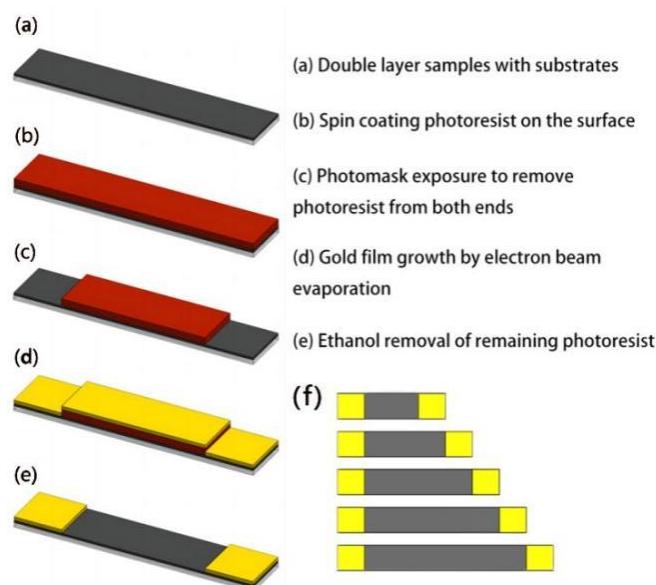


Figure 1. (a)~(e) The process of preparing gold electrodes on the sample surface; (f) Five samples obtained without using channel lengths

2.3. Preparation of gold anode electrode

Gold electrodes were obtained by using the photolithography technique [16, 17], which involved directly attaching a gold film on top of the prepared graphene. The Fig. 1 (a~e) illustrates the process of gold electrode preparation. Firstly, a photoresist is uniformly dropped onto the sample surface consisting of two layers (substrate sub/graphene) and is then rotatively coated to form a 200 nm thick positive photoresist. The photoresist is then soft baked using a heating plate to reinforce it. Afterward, the sample is aligned with the pattern using photomask exposure to remove the photoresist from the two end parts. On the top layer, a 100 nm thick gold film is grown using electron beam evaporation as the electrode layers. [18, 19] Subsequently, the remaining positive photoresist was removed using

ethanol as the debinding solution. In conclusion, five graphene samples with different channel lengths of attached gold electrodes were obtained in Fig. 1 (f).

3. Results And Analysis

3.1. Raman detection

Raman spectroscopy is already one of the main tools for characterizing graphene and can be used to determine the number of layers, doping, defects, and other corresponding properties of graphene [20, 21]. Additionally, for graphene films prepared by different methods, very fine defects can also be characterized using Raman spectroscopy [22]. The D, G, and G' (2D) peaks are distinct characteristic peaks in the Raman spectrum of graphene. The G peak can effectively indicate the number of layers of graphene, while the D and 2D peaks are used to characterize the structural defects or the arrangement of edges and carbon atoms between graphene layers. [23]

Fig.2 (a) shows the Raman spectra of the graphene sample prepared using the CVD method. The G peak is located at approximately 1585 cm^{-1} , and the G' peak is located at approximately 2686 cm^{-1} . The 2D peak exhibits an almost symmetrical perfect Lorentzian peak pattern, with a width of about 34 cm^{-1} at half height. The intensity ratio of the D peak to the G peak (ID/IG) and the full width at half maximum (FWHM) of the G peak are commonly used to characterize the defect density in graphene, as the defect density is proportional to the intensity ratio. Figure b shows the Raman spectrum of FDTD-modified graphene, where the intensities of the two characteristic peaks, the 2D peak and the G peak, are weakened, while the defect peak (D peak) is significantly weakened. This indicates a significant reduction in surface defects on the sample.

The use of Raman spectroscopy for the determination of graphene has a certain level of accuracy, especially for few-layer graphene. In this experiment can be observed that the structure and main composition of graphene remain unchanged after spin-coating with FDTD.

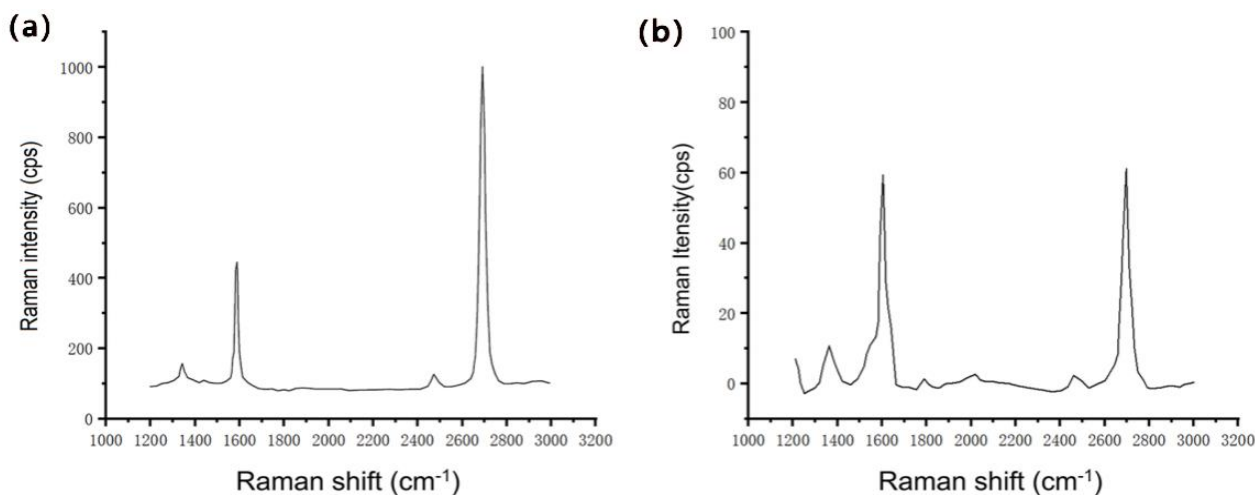


Figure 2. The Raman spectra of the samples before(a) and after(b) the FDTD

3.2. Study on surface wettability of graphene thin films

The self-assembled molecule FDTD reacts with the surface of the sample and forms a regular single-layer molecular membrane on the surface of the sample, which changes the surface structure of the sample, makes the sample hydrophobic and reduces the surface energy of the sample.

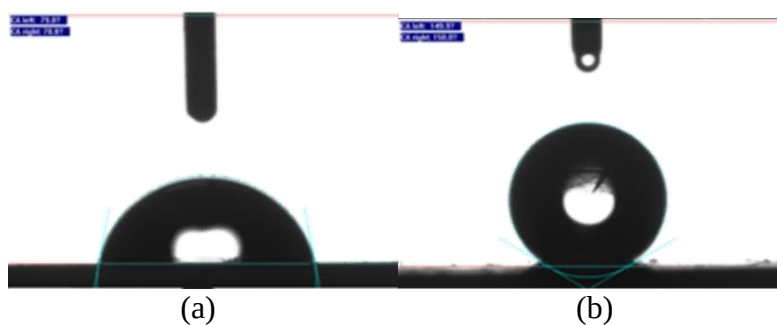


Figure 3. (a) Contact angle of deionized water before reshaping; (b) Contact angle of deionized water after reshaping

According to Owens' two-liquid equation, polar reagent water and non-polar reagent ethylene glycol are selected.

$$\gamma_L(1 + \cos \theta) = 2(\gamma_S^d \cdot \gamma_L^d)^{\frac{1}{2}} + 2(\gamma_S^p \cdot \gamma_L^p)^{\frac{1}{2}} [24] \quad (1)$$

When the base contacts the surface, a contact angle is formed and measured using a contact angle measuring instrument before any base modification, Owens' two-liquid method is shown in Fig. 3. Next, the graphene film surface is modified. The process involves immersing the silicon base in an FDTD solution for a specific duration, followed by getting the silicon wafer, cleaning, and drying it to obtain the FDTD-modified silicon base. The contact angles between the modified substrate and the two reagents were measured using a contact angle measuring instrument, and the average value was obtained from multiple measurements.

Using two-liquid equations, we can calculate the polar component γ_S^d and nonpolar component γ_S^p of surface energy of graphene film. Finally, the polar component and nonpolar component are added to obtain the surface energy of solid.

We will drop the deionized water to the surface of the unmodified graphene film sample, and use the instrument to measure the contact angle of the sample when the balance is achieved. Similarly, drip the ionized water to the surface of the dry samples modified by the FDTD, and use the instrument to measure the contact angle of the sample when it is balanced. We repeat this process for samples modified at different times, measuring their contact angles and obtaining the average value through multiple measurements. The measurement results are depicted in Fig. 4.

Based on the analysis, it is believed that the hydrophobic angle of the sample before coating is 75.36° , indicating that the base itself is hydrophilic. After processing, the actual contact angle of the sample is close to 0 degrees, which is super affordable. However, after being modified by the FDTD, the contact angle of the sample is significantly increased. The static contact angle of the sample surface, measured at different processing times, exceeds 140° , indicating that the surface has become hydrophobic. This transformation from hydrophilicity to hydrophobicity is attributed to the interaction between FDTD molecules and the hydroxyl groups present on the surface of the graphene sample. Because a hydrolyzed csontraction reaction occurs between the FDTD molecules on the surface of the graphene sample, the surface forms a regular discharge, which makes the sample hydrophobic. The total process of the sample reaction to the FDTD molecular reaction realizes the transformation from hydrophilicity to hydrophobicity. Through the formula, using the contact angle between known samples and water and ethylene glycol, you can calculate the free energy of the sample surface modified by the FDTD. In the analysis, it is determined that the free energy of the graphene film sample surface, which has not been treated by FDTD, is approximately $29.72 \text{ mJ}\cdot\text{m}^{-2}$.

The surface energy of graphene film samples versus FDTD reaction time is shown in Fig. 3 (c). The analysis believes that when it is treated without FDTD, the free energy of the graphene film sample surface can be $29.72 \text{ mJ}\cdot\text{m}^{-2}$; When the sample surface is just dripped on FDTD, the surface free energy can be $32.31 \text{ mJ}\cdot\text{m}^{-2}$. When the action time respectively are 1h, 2h, 4h, 8h, 12h, 24h, the free energy can be $30.60 \text{ mJ}\cdot\text{m}^{-2}$, $25.06 \text{ mJ}\cdot\text{m}^{-2}$, $18.49 \text{ mJ}\cdot\text{m}^{-2}$, $21.53 \text{ mJ}\cdot\text{m}^{-2}$, $38.10 \text{ mJ}\cdot\text{m}^{-2}$, 12.32

$\text{mJ}\cdot\text{m}^{-2}$, an increase of 2.96%, a decrease of 15.68%, a decrease of 37.79%, a decrease of 27.56%, an increase of 28.20%, a decrease of 58.55%, compared to not modified. The free energy of the specimen surface is closely related to the time of the FDTD reaction. As the samples reacted with FDTD self-assembly for some time, the surface energy of the coated specimens was significantly lower than that of the uncoated specimens.

To further understand the optimal reaction time for achieving the lowest surface free energy, time gradient experiments were conducted. These experiments involved changing the reaction time of the graphene films with FDTD, while measuring the surface energy at different points in the reaction. The results depicting the relationship between the surface energy of the graphene film and the reaction time are presented in Fig. 4 (d).

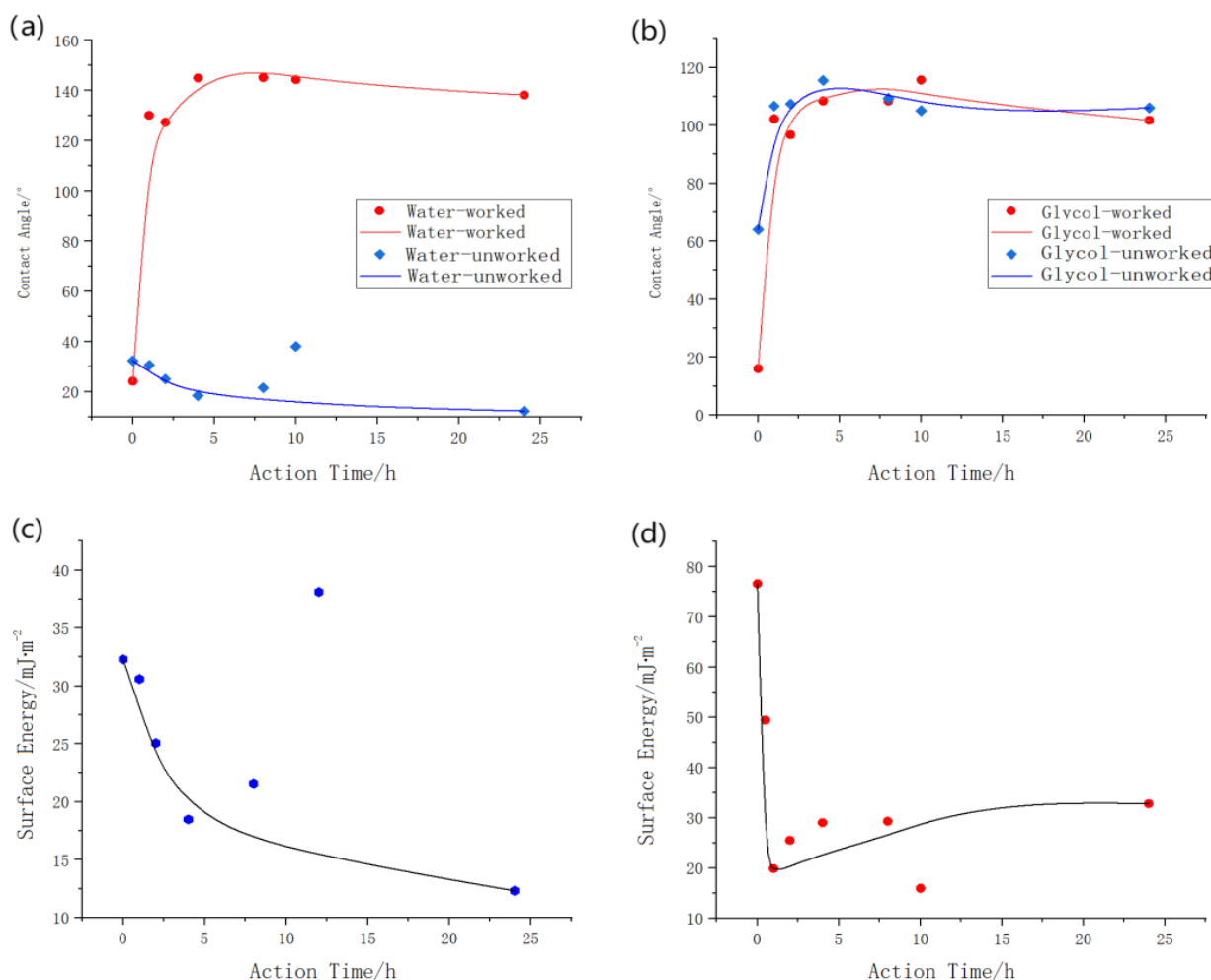


Figure 4. (a)Polar reagent water to measure contact angle; (b) non-polar reagent ethylene glycol to measure contact angle; (c), (d)Surface energy of graphene films as a function of reaction time

It was analyzed that the surface energy of the untreated graphene specimen was $36.38382 \text{ mJ}\cdot\text{m}^{-2}$ the free energy of the surface of the specimen when freshly coated was $76.61118 \text{ mJ}\cdot\text{m}^{-2}$. The uncoated graphene is about 0.34 nm thick, and the hydrophobic properties are greatly changed this time compared to the surface of a titanium film coated with a layer about 1 nm thick. The free energy was $19.89465 \text{ mJ}\cdot\text{m}^{-2}$ for a reaction time of 1h, which was 45.32% lower compared to the uncoated one; The free energy was $25.53085 \text{ mJ}\cdot\text{m}^{-2}$ for a reaction time of 2h, which was 29.83% lower compared to the uncoated one; The free energy was $29.04612 \text{ mJ}\cdot\text{m}^{-2}$ for a reaction time of 4h, which was 20.17% lower compared to the uncoated one. The free energy of the specimen surface is lowest at this time; The free energy was $29.33762 \text{ mJ}\cdot\text{m}^{-2}$ for a reaction time of 8h, which was 19.37% lower compared to the uncoated one; The free energy was $15.95723 \text{ mJ}\cdot\text{m}^{-2}$ for a reaction time of

10h, which was 56.14% lower compared to the uncoated one; The free energy was 32.84367 mJ·m⁻² for a reaction time of 24h, which was 9.73% lower compared to the uncoated one.

Based on these results, it can be concluded that when the graphene specimen reacts with FDTs molecules, the surface energy generally decreases compared to the uncoated specimen. The optimal reaction time is between 10-24h, with the most ideal reaction occurring at 10h, which results in the lowest surface energy.

3.3. Electrical property test

The resistance of graphene specimens was investigated to understand how it changes with the degree of binding between the specimens and FDTs self-assembled molecules. To achieve this, the reaction time of graphene films with FDTs was varied, and the average resistance of the graphene films was measured at different reaction times.

The resistance measurements were conducted by attaching a micro-nano-scale probe stage to the graphene specimens, and the resulting data were curve-fitted. We use the four-probe method with the modified graphene on the determined position according to the method, set the two farthest probes as current inflow and current outflow, and then measure the potential difference between the two nearest probes and thus the result.

For trench 1, the average length of the trench was 201.71nm. The resistance of the graphene specimen was measured without FDTs treatment as 173.13 Ω. The resistance of the specimen immersed for 1 h was 81.25 Ω; the resistance of the specimen immersed for 2 h was 26.80 Ω; the resistance of the specimen immersed for 4 h was 16.45 Ω; the resistance of the specimen immersed for 8 h was 6.66 Ω; the resistance of the specimen immersed for 10 h was 1.22 Ω; and the resistance of the specimen immersed for 24 h was 1.52 Ω. The other four trenches are in a similar situation. We summarized the data for the five trenches.

The average resistance of the graphene specimens measured without FDTs is 168.81 Ω. The resistance of the specimen immersed for 1 h is 99.66 Ω, which is 40.96% lower than that of the unsoaked specimen; The resistance of the specimen immersed for 2 h is 34.37 Ω, which is 79.64% lower than that of the unsoaked specimen; The resistance of the specimen soaked for 4 h was 12.48 Ω, which was 92.61% lower than that of the unsoaked specimen; the resistance of the specimen soaked for 8 h was 5.24 Ω, which was 96.90% lower than that of the unsoaked specimen; the resistance of the specimen soaked for 10 h was 1.30 Ω, which was 99.23% lower than that of the unsoaked specimen; the resistance of the specimen soaked for 24 h was 1.74 Ω, which was 98.97% lower than that without soaking.

According to the carrier mobility equation [25], the lower the resistance, the higher the carrier mobility as the reaction time increases:

$$\mu = \frac{L}{neRS} \quad (2)$$

The multi-terminal electrical measurements show an electron mobility of ~5,740 cm²/V·s at a carrier density of ~5 × 10¹² cm⁻² [26] and an average line resistance of 1.3 Ω for a reaction time at the 10 h in Fig. 5 (c).

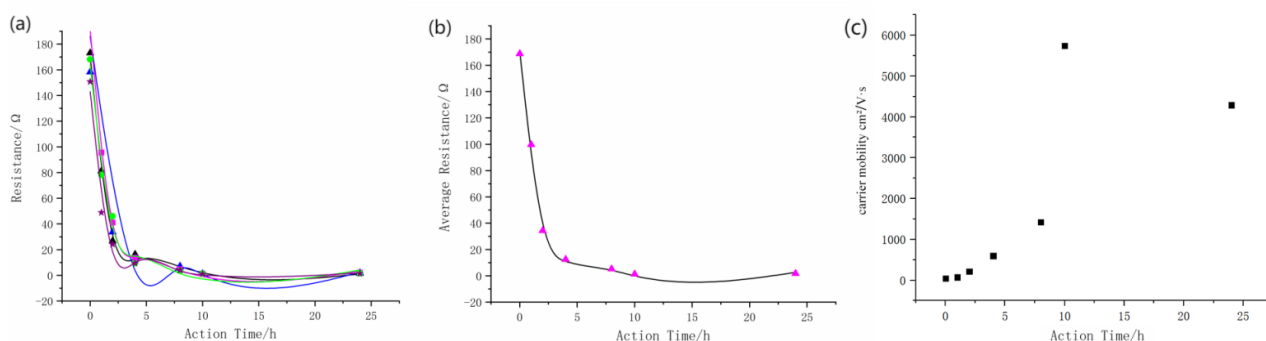


Figure 5. (a) fitting analysis of sample resistance data of different 5 channels; (b) fitting analysis of the average linear resistance data of graphene sample; (c) Analysis of graphene carrier mobility with time

This process was repeated multiple times for graphene specimens with the same channel, but immersed in FDTS for different reaction times. The average line resistance of the graphene specimens, obtained from several repetitions of the experiment, is presented in Figure 5 (b). The results indicate that the surface conductivity of graphene was increased significantly, by nearly 130 times, after reacting with FDTS. The resistance data acquired from the probe measurements show that, within a certain time range, the longer the Specimens are immersed in FDTS, the lower their resistance becomes. This suggests that the reaction between the graphene specimens and FDTS is more thorough, resulting in reduced resistance. Furthermore, it can be observed that the resistance of treated graphene decreases as the binding degree of the surface modification molecules to graphene increases. The most favorable reaction effect occurs when the specimen is immersed in FDTS for a duration of 10-24 hours, and the carrier mobility also reaches a high value of $\sim 5,740 \text{ cm}^2/\text{V}\cdot\text{s}$.

4. Conclusion

In this paper, we employ monolayers of graphene fabricated through CVD and subject them to surface modification using FDTS with varying reaction durations with the range of 10 to 24 hours recommending. To comprehensively characterize the modified graphene, we adopt a multi-faceted approach encompassing microscopic inspections, Raman spectroscopy, contact angle measurements, and probe electrical tests. Our investigation yields noteworthy findings. The application of FDTS to graphene leads to a discernible reduction in surface defects, thereby diminishing the graphene surface energy from an initial value of 28.21 mJ/m^2 to an exceptionally low 0.79 mJ/m^2 , with the notable reduction in both polar and non-polar components. Subsequent measurements of the treated graphene demonstrate a pronounced decline in average resistance, ascribing this change to the strengthened binding of the modified molecular layer to the graphene substrate. Resistance decreases from an initial value of $168.81 \text{ }\Omega/\mu\text{m}$ to a mere $1.29 \text{ }\Omega/\mu\text{m}$., which leads to a nearly 130-fold increase in surface conductivity, provides a new avenue for research and innovation in this area.

References

- [1] Venderbos, Jörn WF, and Rafael M. Fernandes. "Correlations and electronic order in a two-orbital honeycomb lattice model for twisted bilayer graphene." *Physical Review B* 98.24 (2018): 245103.
- [2] Iqbal A K M A, Sakib N, Iqbal A K M P, et al. Graphene-based nanocomposites and their fabrication, mechanical properties and applications [J]. *Materialia*, 2020, 12: 100815.
- [3] Yousefi, Nariman, et al. "Environmental performance of graphene-based 3D macrostructures." *Nature nanotechnology* 14.2 (2019): 107 - 119.
- [4] Chen Z, Narita A, Müllen K. Graphene nanoribbons: on-surface synthesis and integration into electronic devices [J]. *Advanced Materials*, 2020, 32 (45): 2001893.
- [5] Akinwande, Deji, et al. "Graphene and two-dimensional materials for silicon technology." *Nature* 573.7775 (2019): 507 - 518.
- [6] Ono, Masaaki, et al. "Ultrafast and energy-efficient all-optical switching with graphene-loaded deep-subwavelength plasmonic waveguides." *Nature Photonics* 14.1 (2020): 37 - 43.
- [7] Guo H, Garro-Hernandorena A, Martínez-Galera A J, et al. Lateral Heterostructures of Graphene and h-BN with Atomic Lattice Coherence and Tunable Rotational Order [J]. *Small*, 2023, 19 (17): 2207217.
- [8] Tiwari, Santosh K., et al. "Graphene research and their outputs: Status and prospect." *Journal of Science: Advanced Materials and Devices* 5.1 (2020): 10 - 29.
- [9] Deng, Bing, Zhongfan Liu, and Hailin Peng. "Toward mass production of CVD graphene films." *Advanced Materials* 31.9 (2019): 1800996.

- [10] Deng B, Liu Z, Peng H. Toward mass production of CVD graphene films [J]. *Advanced Materials*, 2019, 31 (9): 1800996.
- [11] Wang, Huaping, et al. "Primary nucleation-dominated chemical vapor deposition growth for uniform graphene monolayers on dielectric substrate." *Journal of the American Chemical Society* 141. 28 (2019): 11004 - 11008.
- [12] Sun J, Zhao J, Chen Y, et al. Macro-micro-nano multistage toughening in nano-laminated graphene ceramic composites [J]. *Materials Today Physics*, 2022, 22: 100595.
- [13] Wang L, Urbas A M, Li Q. Nature-inspired emerging chiral liquid crystal nanostructures: from molecular self-assembly to DNA mesophase and nanocolloids [J]. *Advanced Materials*, 2020, 32 (41): 1801335.
- [14] Chu, Zhenming, et al. "Smart superhydrophobic films with self-sensing and anti-icing properties based on silica nanoparticles and graphene." *Advanced materials interfaces* 7.15 (2020): 2000492.
- [15] Li, Jinjin, et al. "Molecular origin of super lubricity between graphene and a highly hydrophobic surface in water." *The journal of physical chemistry letters* 10.11 (2019): 2978 - 2984. S. C. Xu. *Studies of Graphene on Preparation, Characterisation, Optoelectronic Properties and Applications* [D]. Shandong Normal University, 2014.
- [16] Tanwar A, Gandhi H A, Kushwaha D, et al. A review on microelectrode array fabrication techniques and their applications [J]. *Materials Today Chemistry*, 2022, 26: 101153.
- [17] Hondred J A, Johnson Z T, Claussen J C. Nanoporous gold peel-and-stick biosensors created with etching inkjet maskless lithography for electrochemical pesticide monitoring with microfluidics[J]. *Journal of Materials Chemistry C*, 2020, 8 (33): 11376 - 11388.
- [18] Mishra Y K, Mohapatra S, Kabiraj D, et al. Growth of Au nanostructures by annealing electron beam evaporated thin films [J]. *Journal of Optics A: Pure and Applied Optics*, 2007, 9 (9): S410.
- [19] Hasna K, Anju K S, Jayaraj M K. Photonic and optoelectronic applications of graphene: Applications of graphene in surface-enhanced Raman scattering[M]//Recent Advances in Graphene and Graphene-Based Technologies. Bristol, UK: IOP Publishing, 2023: 10 - 1 - 10 - 24.
- [20] Hulman M. Raman spectroscopy of graphene[M]//Graphene. Woodhead Publishing, 2021: 381-411.
- [21] Li Z, Deng L, Kinloch I A, et al. Raman spectroscopy of carbon materials and their composites: Graphene, nanotubes and fibres [J]. *Progress in Materials Science*, 2023: 101089.
- [22] Merlen A, Buijnsters J G, Pardanaud C. Raman spectroscopy characterization of carbon materials: From graphene to all-carbon heterostructures [J]. *All-carbon composites and hybrids*, 2021: 317 - 346.
- [23] Orlando A, Franceschini F, Muscas C, et al. A comprehensive review on Raman spectroscopy applications [J]. *Chemosensors*, 2021, 9 (9): 262.
- [24] Owens D K, Wendt R C. Estimation of the surface free energy of polymers [J]. *Journal of applied polymer science*, 1969, 13 (8): 1741 - 1747.
- [25] Gosling J H, Makarovskiy O, Wang F, et al. Universal mobility characteristics of graphene originating from charge scattering by ionised impurities [J]. *Communications Physics*, 2021, 4 (1): 30.
- [26] Sonntag J, Watanabe K, Taniguchi T, et al. Charge carrier density dependent Raman spectra of graphene encapsulated in hexagonal boron nitride [J]. *Physical Review B*, 2023, 107 (7): 075420.