

## Optimizing Atmospheric Ion Harvesting Electrodes with Graphene for Clean Energy Generation Based on Capacitive Properties and Energy Storage

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### Abstract

The atmosphere is rich in positive ions, rendering it electrically more positive than the Earth's surface. This characteristic presents the atmosphere as a potential source of renewable energy through ion harvesting. This study harnesses the electrical properties by optimizing ion harvesting electrodes using pristine graphene and graphene-Au thin films to generate clean electricity. Research methods included Raman Spectroscopy and Cyclic Voltammetry (CV) to assess the surface characteristics and capacitance of the graphene samples, along with laboratory-scale ion harvesting simulations to evaluate the energy data produced in the ion harvesting process. The samples used in this study were identified as bilayer graphene, as confirmed by Raman Spectroscopy. CV testing yielded capacitance values of 0.40288 F for pristine graphene and 0.44879 F for graphene-Au samples. According to ion harvesting simulations, graphene-Au generated approximately 6.8 times more energy than pristine graphene and five times more energy than copper alone. The respective energy outputs for graphene-Au, pristine graphene, and pure copper were 1.376 mW, 1.157 mW, and 0.374 mW. These results demonstrate that adding a graphene layer to the atmospheric ion-harvesting electrode can optimize the electricity generation process.

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## INTRODUCTION

The modern lifestyle is heavily reliant on electricity consumption, as demonstrated by the national electricity consumption data in Indonesia, which reached 348 TWh in 2022 (Directorate General of Electricity, Ministry of Energy and Mineral Resources, 2023). Unfortunately, a significant portion of Indonesia's electricity is still generated by fossil fuels, accounting for 85% of thermal power plants (Directorate General of Electricity, Ministry of Energy and Mineral Resources, 2023). Consequently, Indonesia contributes substantially to CO<sub>2</sub> emissions. The 2023 Ember Climate Watch Report revealed that Indonesia ranks 9th in CO<sub>2</sub> emissions, with 192.7 million tons of CO<sub>2</sub> originating from the energy sector (Mutiarra, 2023). The reliance on fossil fuels, if left unchecked, will adversely affect Indonesia's environment and public health. In response, Jisman Hutajulu, from the Directorate General of Electricity, suggests energy sector mitigation measures, including the implementation of new and renewable energy, energy efficiency applications, the adoption of low-carbon fuels, and the use of clean technologies (EBKTE Public Relations, 2023).

Numerous alternatives exist for new and renewable energy generation, with atmospheric ions being one of them. The atmosphere harbors an abundance of positive ions ready to be harvested and converted into electrical energy. Due to air pollution, cosmic radiation, and the Earth's intrinsic negativity, the density of positive ions is 10-20% higher than negative ions (Liu, 2017). The quantity of ions increases during adverse weather conditions, such as thunderstorms and other atmospheric situations. Consequently, the sky tends to possess a more positive electric field compared to the Earth's surface. Utilizing ion-harvesting technology involves connecting conductor electrodes stored in the sky at specific altitudes, generating electrical energy by allowing current flow in the cable connection due to the electrode's effort to neutralize the positive ions in the sky (Feynman *et al.*, 2014). Typically, using copper electrodes can produce electrical power of 0.481 watts (Hussain *et al.*, 2021). With the introduction of graphene nanomaterials and a capacitor needle model as an ion harvester, there is potential for increased power generation and energy charging efficiency.

Graphene is a two-dimensional nanocarbon material with a single-atom thickness that exhibits excellent properties such as mechanical strength, electrical and thermal conductivity, elasticity, transparency, capacitance, and a high surface area

(Vestince *et al.*, 2021). With these characteristics, it is highly feasible to generate clean and relatively stable energy that is not weather-dependent and does not produce any waste, overcoming the shortcomings of solar and nuclear power. Solar energy faces a limitation in its dependency on sunny weather (Igini, 2023). Proposed solutions include using molten salts for daytime heat storage and deploying solar panels on high-flying balloons to capture energy above clouds (Sheikh, 2016; Biello, 2009). However, these solutions are not easily applicable at the household level (Liu *et al.*, 2017). In contrast, nuclear power generates waste necessitating specialized management (Igini, 2023). The primary strategies for nuclear waste management involve recycling used nuclear fuel and direct disposal in underground repositories, but these approaches pose technical challenges, social issues, and may erode trust in responsible institutions (Ramana, 2018).

In this study, we optimized ion harvester electrodes with graphene to generate clean electrical energy. Therefore, the objective of this research is to determine and analyze the surface characteristics and electrical capacitance of pristine graphene through Raman spectra and the current-voltage curve characteristics of CV. Additionally, we aimed to analyze the electrical characteristics of graphene electrodes using a capacitor model by measuring the power and efficiency of the charging and discharging times as a simulation of ion capture from the atmosphere on a laboratory scale. The characterization tests conducted in this research aimed to identify the best graphene sample type and simulate the early stages of ion-to-electricity conversion by storing energy in a capacitor. It is hoped that this study will inspire further research and innovation in the field of atmospheric ion-based electrical energy generation, enabling rapid mitigation of renewable energy actions and the development of sustainable clean energy sources to reduce the impact of CO<sub>2</sub> emissions from conventional energy generation.

## METHODS

This research was conducted over a period of 4 months, from July to October 2023, at the following locations: the Material and Instrumental Physics Laboratory of the Faculty of Mathematics and Natural Sciences, Universitas Gadjah Mada (UGM), and the Atomic and Nuclear Physics Laboratory of the Faculty of Mathematics and Natural Sciences, UGM. The research utilized various tools and materials, including scissors, Chemical Vapor Deposition (CVD) equipment,

Raman Spectrophotometer (LabRAM HR Evol), CV equipment (Corrtest), sandpaper sheets, tweezers, measuring glass, spatula, beaker glass, graduated flask, 3.7 V Li-ion battery charger, solder, copper foil, pristine graphene, graphene-Au, acrylic capacitor equipment, spacer, bolts, separator paper, 1M KCl solution, deionized water, acrylic simulation equipment, alligator clip cables, mini power switch, cables, 15 kV high-voltage generator, multimeter, voltage sensor, resistor, 3.7 V Li-ion battery, black tape, and tin.

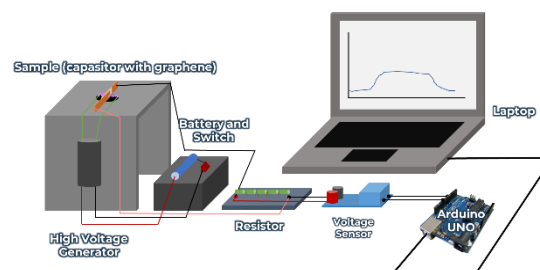
The samples were initially prepared by synthesizing pristine graphene using the CVD method, a technique for depositing materials as thin films onto substrates from vapor species through chemical reactions (Saeed *et. al.*, 2020). This method was employed to obtain pristine graphene with a thickness of only a few layers without the presence of oxide bonds. During this process, graphene was grown on a thin copper substrate with an area of  $2 \times 1 \text{ cm}^2$ . After the successful synthesis of graphene, one of the samples was doped with gold (Au) using the spin-coating technique for 120 seconds.

The data collected in this research includes the results of Raman spectroscopy tests and CV tests to determine the surface characteristics and capacitance values of graphene layer samples on copper foil. Laboratory-scale atmospheric ion harvesting simulations were also conducted to obtain data on the energy generated by the electrode samples. Raman spectroscopy is a spectroscopic technique based on the inelastic scattering of monochromatic light, causing molecular deformations by an electric field (Sunu *et al.*, 2019). The use of Raman spectroscopy for sample testing is also included in the characterization of optical properties, utilizing the interaction of light with a material to determine the number of layers (Dwi *et al.*, 2021). In this research, Raman spectroscopy was focused on graphene samples that had been attached to their substrates to determine the analysis of their surface characteristics, namely surface defect levels and the number of graphene layers, which can be discerned from the comparison of Raman spectrum intensity values.

CV is a technique used to understand and characterize the reduction-oxidation processes, stability, and effective surface area of electrodes for biosensing. In the testing process, graphene material can act as a transducer converting ions into measurable electrons. Typically, CV testing consists of three electrodes: the working electrode (WE), the reference electrode (RE), and the counter electrode (CE). During the testing process, voltage was applied to the WE back and forth for a certain

number of cycles, allowing the WE current to be measured. The potential at which the current reaches its peak is known as the peak potential ( $E_p$ ), which can be used to analyze electrochemical reversibility (Ramli *et al.*, 2018). In this study, CV tests were conducted on electrodes assembled as supercapacitors to analyze the capacitance and conductivity of the samples.

In the laboratory-scale ion harvesting simulation phase, the design and assembly of the ion harvesting simulation device were implemented, as illustrated in Figure 1. One of the components of this simulation device is a 15 kV high-voltage generator that is connected to the test sample through two electrodes with different potentials. When the device is in operation, it generates minor plasma sparks accompanied by sounds, resembling the conditions of Earth's atmosphere with an electric field gradient, and the presence of high voltage leading to ionization processes in the air molecules around the plasma. This simulates the analogy of an atmosphere dominated by positive ions. Through this setup, the device was configured to measure current and voltage values under various resistance conditions. This allowed for an analysis to determine the generated power, as well as the charging and discharging time constants of the capacitor.



**Figure 1.** Diagram of atmospheric ion harvesting simulation data acquisition

The data analysis conducted in this research is as follows. In the Raman spectroscopy test, the spectra of the D (this band signifies a ring breathing mode originating from  $sp^2$  carbon rings), 2D (it manifests as a distinct peak at approximately  $1587 \text{ cm}^{-1}$  in the graphene spectrum, representing an in-plane vibrational mode of the  $sp^2$  hybridized carbon atoms forming the graphene sheet), and G bands (it arises from a two-phonon lattice vibrational process) for both pristine graphene and graphene-Au samples were observed. Subsequently, the intensity ratios were determined, specifically ID/IG to characterize the surface defects of graphene and I2D/IG to ascertain the number of graphene layers (David, 2016). Data analysis for the Raman spectrum was

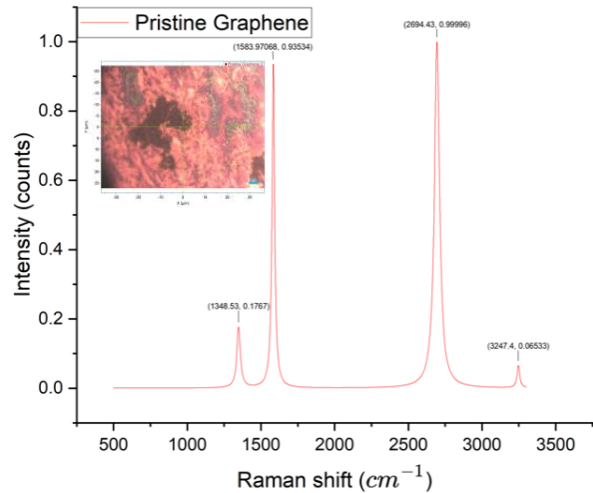
conducted using the OriginPro software. In the CV test, the current-voltage curve obtained was observed, followed by regression analysis to determine the resistance and conductivity values, subsequently obtaining the capacitance value through area analysis. Data analysis of the current-voltage data was conducted using the CS Studio 6 application. In the laboratory-scale ion harvesting simulation, the time constant value was sought to determine how quickly the capacitor could charge and discharge its load. The current and power values that could be generated were also calculated by fitting exponential curves to the voltage-time plot data. Data analysis for this was performed using the OriginPro software.

## RESULTS AND DISCUSSION

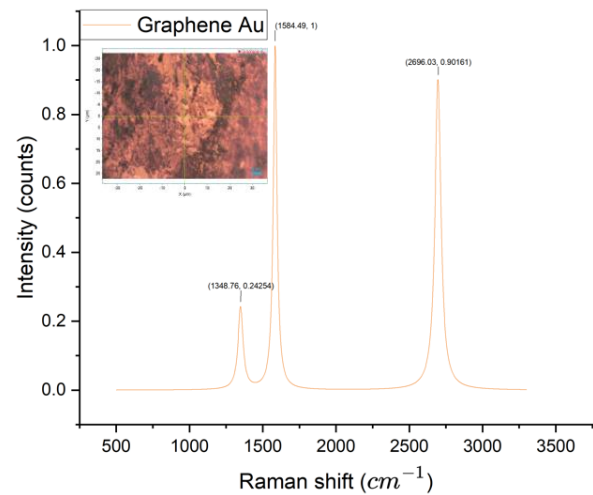
### A. Raman Spectroscopy Characterization

Following the successful synthesis of graphene samples using the CVD method on copper foil sheets, and with some samples doped with Au, both samples underwent Raman testing using the LabRAM HR Evol instrument at a wavelength of 523 nm. Figure 2 illustrates the Raman spectrum profiles and surface appearance for each sample. Slight color variations are observed between pristine graphene and graphene-Au. The graphene-Au sample exhibits a more golden hue, while the pristine graphene appears more red, indicating a relatively thick gold particle doping in the sample. It is notable that the 2D spectrum in pristine graphene is higher compared to the spectrum of graphene-Au, an outcome attributed to the sensitivity of the 2D spectrum to sample thickness.

Based on the I<sub>2D</sub>/I<sub>G</sub> values, the pristine graphene exhibited a value of 1.07, while the graphene-Au had a value of 0.9. These values indicate that both samples are graphene bilayers, consisting of two layers of graphene. Additionally, the I<sub>D</sub>/I<sub>G</sub> values, which represent the defect levels, were relatively low for both samples, with 0.18 for pristine graphene and 0.24 for graphene-Au. The increase in defect values in graphene-Au is attributed to the introduction of gold elements on the graphene surface. These values indicate that the electrode samples coated with graphene are in good condition with minimal defects. A comparison diagram of I<sub>D</sub>/I<sub>G</sub> and I<sub>2D</sub>/I<sub>G</sub> values, as well as the full-width at half-maximum (FWHM), is presented in Figure 3.



(a)

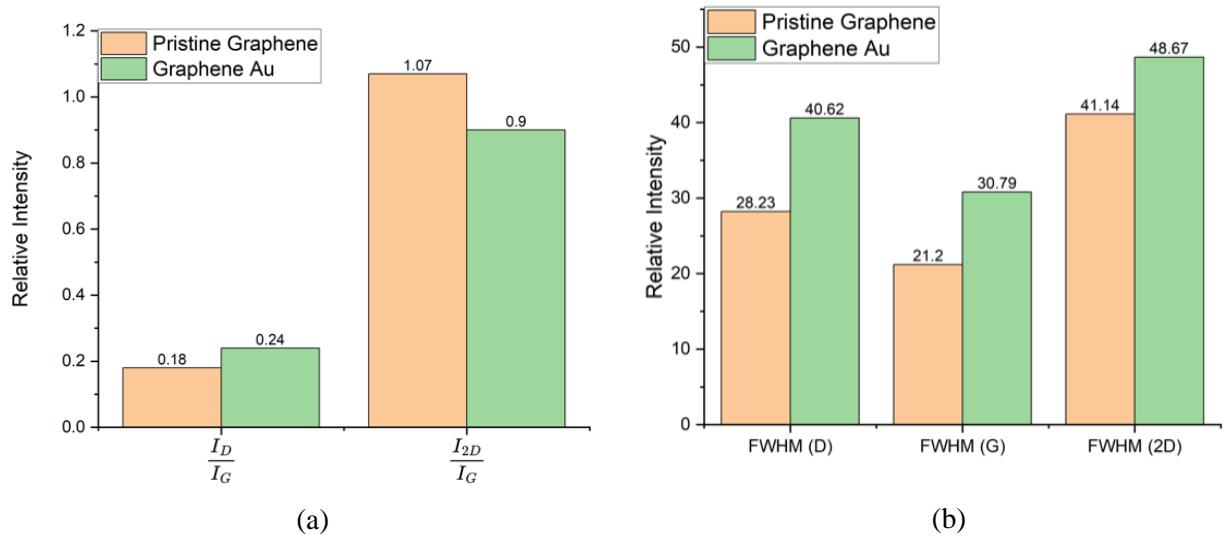


(b)

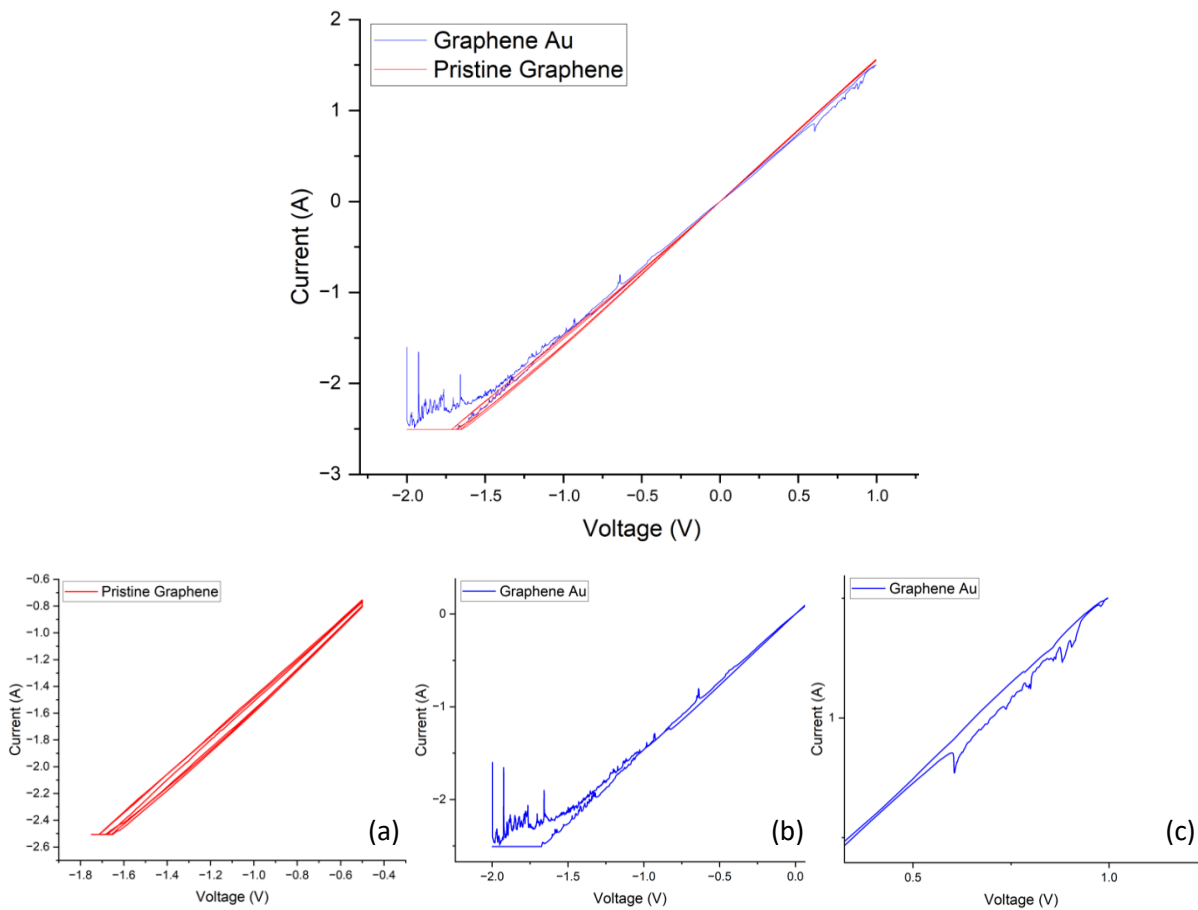
**Figure 2.** Raman spectra along with their surface appearance of (a) pristine graphene and (b) graphene-Au

### B. Cyclic Voltammetry Characterization

The CV tests were conducted at the UGM Material Physics and Instrumentation Laboratory using the Corrtest device. Prior to characterization, the samples were prepared in a capacitor model and inserted with a separator containing a 1 M KCl solution between the two electrodes. The testing was performed at a scan rate of 50 mV/s from -2 to 1 V. The results that presented in Figure 4 yielded linear I-V characteristic graphs indicating the conductive nature of the samples. The computed values for resistivity, conductivity, and capacitance are presented in Table 1.



**Figure 3.** (a)  $I_D/I_G$  and  $I_{2D}/I_G$  values (b) their FWHM values



**Figure 4.** CV test results of the I-V characteristic curve (a) cycle area at negative voltage of pristine graphene (b) cycle area at negative voltage of graphene-Au (c) cycle area at positive

The I-V characteristic curve obtained does not closely align with the typical CV curve for copper electrode coated with pristine graphene. This discrepancy may result from suboptimal data acquisition methods. The linear I-V characteristic curve suggests the presence of Ohmic contacts. The wide voltage range and high scan rate are estimated to cause the cycle area to be less clearly visible. Additionally, data acquisition using a supercapacitor assembly, rather than using electrodes in an electrolyte solution, is one of the contributing factors. The difficulty in achieving a supercapacitor arrangement in the form of an electrode with a separator (dielectric material) in between to maintain a very close distance approaching a vacuum also contributes to the lack of clarity in the cycle curve. It is highly recommended to employ improved data acquisition methods for future research.

**Table 1.** Resistivity, Conductivity, and Capacitance Values of the Samples

Parameter	<i>Graphene</i>	<i>Graphene-Au</i>
Conductivity (S/m)	147.1681289	145.3982225
Resistivity (ohm)	0.679494947	0.687766317
Capacitance (F)	0.40288	0.44879

Although the cyclic curves may not be very distinct, conductivity and resistivity calculations can still be obtained from the curve's gradient. Additionally, capacitance values can be determined by calculating the area under the curve. The results of the calculations in Table 1 indicate that the capacitance of both graphene samples on copper foil approximates that of a conventional supercapacitor (1-100 F). These values are significantly better than typical capacitor capacitances, which are usually within the range of picofarads to microfarads.

The conductivity values obtained based on the gradient of the linear curve fitting are relatively close to those of previous graphene research, specifically 106 S/m (Vestince *et al.*, 2021). This suggests that the graphene used in this study exhibits good quality. This is further supported by the relatively low resistivity values. Comparing the two graphene samples, Graphene-Au exhibits superior capacitance compared to pristine graphene. However, pristine graphene has higher conductivity

than Graphene-Au. This difference could be attributed to the additional gold element in the Graphene-Au sample, which may hinder electron mobility and consequently reduce its conductivity.

### C. Laboratory-Scale Atmospheric Simulation

This simulation was conducted at the UGM Atomic and Nuclear Physics Laboratory using an assembly of a 15 kV high-voltage generator (HV) and data acquisition tool, including a voltage sensor and Arduino. The simulation involved arranging copper layers into a capacitor shape (electrode-separator-electrode), with the electrodes consisting of copper foil sheets and filter paper used as a separator. The HV generator electrode, serving as the high-voltage output, was placed approximately 1 cm from the capacitor situated in the middle. Data collection involved turning on the HV generator for 20 seconds with a Li-ion 4V battery power source, followed by voltage measurement during that period. The results of the analysis, represented by the exponential fitting curve of the capacitor charging and discharging process, are presented in Figure 5, while the calculated results are displayed in Figure 6.

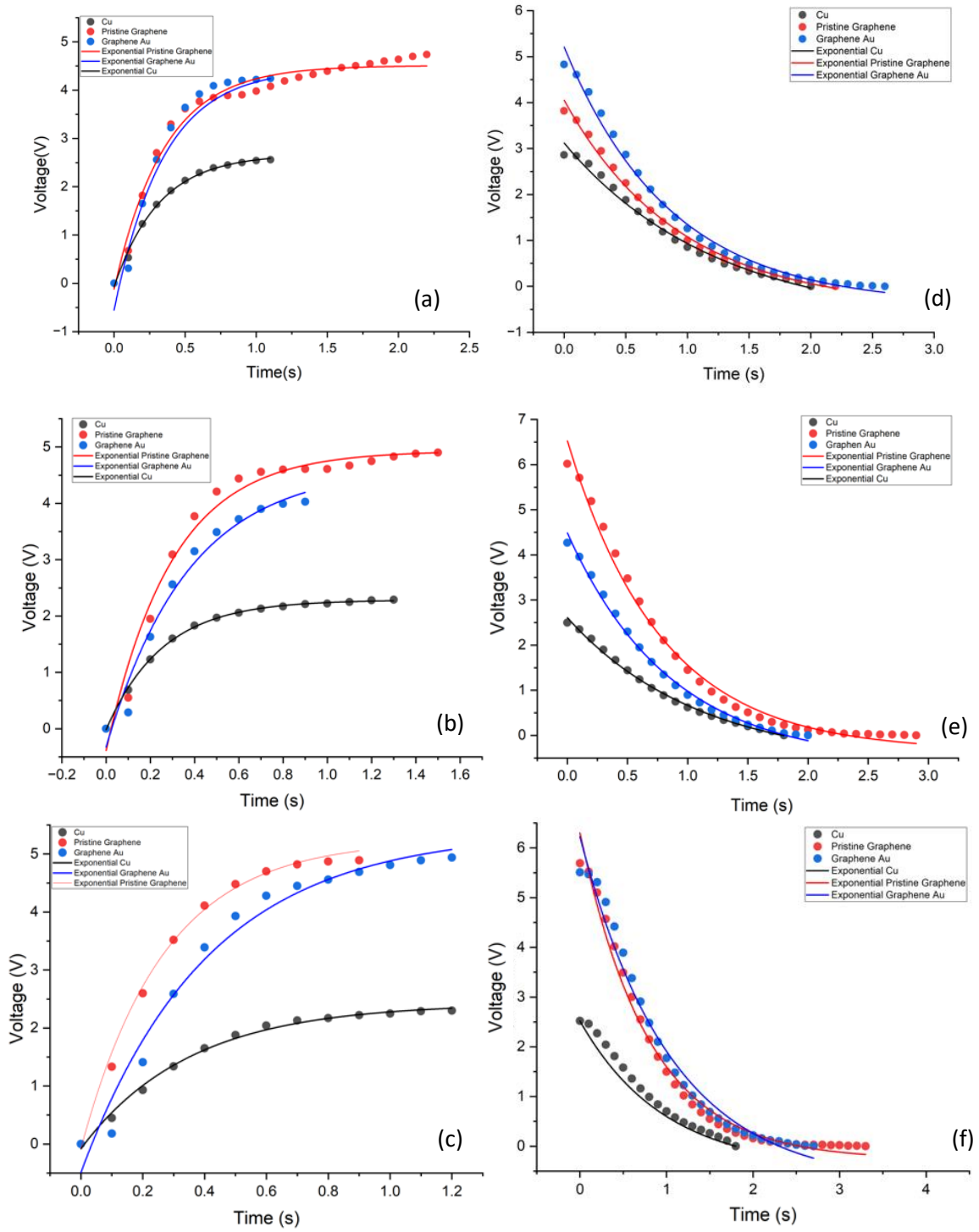
The voltage data measured by the voltage sensor every 0.1 seconds were fitted using the voltage charging capacitor model based on its theoretical equation, which is

$$V(t) = V_o \left( 1 - e^{-\frac{\Delta t}{RC}} \right) \quad (1)$$

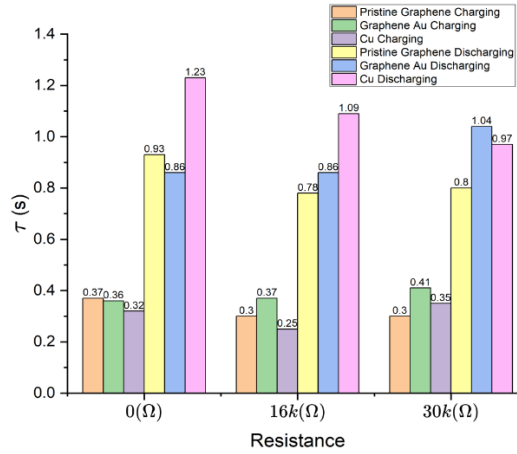
where  $V_o$  is the initial voltage applied,  $\Delta t$  is the time difference between data points, and  $RC$  is the product of resistance and capacitance, which has a value equal to the time constant ( $\tau$ ). The data fitting for the capacitor discharge was done using the model from the following equation.

$$V(t) = V_o e^{-\frac{\Delta t}{RC}} \quad (2)$$

Based on the curve trends in Figure 5, it is evident that the voltage generated by pure graphene samples (red) tends to be higher compared to the other two samples. However, the graphene-Au sample (blue) demonstrates a faster charging time compared to the other two samples. Meanwhile, the copper sample (black) consistently exhibits the lowest voltage across all measurement variables.



**Figure 5.** Laboratory-scale simulation results curve (a) Charging without resistor (b) Charging with  $R = 16k\Omega$  (c) Charging with  $R = 30k\Omega$  (d) Discharging without resistor (e) Discharging with  $R = 16k\Omega$  (f) Discharging with  $R = 30k\Omega$

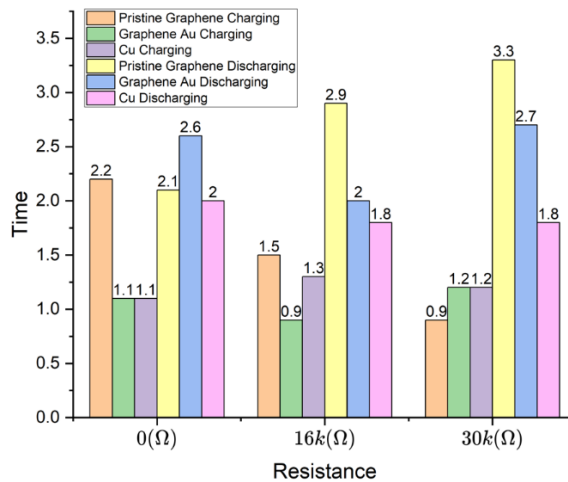


**Figure 4.** Comparison of time constant ( $\tau$ ) values

The time constant values shown in Figure 6 indicate how long or quickly it takes to charge or discharge the capacitor. A favorable value signifies a fast charging time and a long discharge time, indicating that the capacitor can quickly harvest ions and store them for a longer period. The copper sample exhibited the fastest charging time in the circuit without resistance and  $R=16\text{ k}\Omega$ . This was due to the generated voltage not being excessively high, leading to a quicker attainment of voltage saturation. Meanwhile, in the circuit with higher resistance, i.e.,  $R=30\text{ k}\Omega$ , pristine graphene demonstrated the fastest charging time among the two others. When compared to other resistance variations, the charging time of pristine graphene tended to remain unchanged. Additionally, an increasing time constant trend was observed with the increasing resistance in the graphene-Au sample. Concerning the discharge data, the longest discharge time was found for the copper sample. Between the two graphene samples, Graphene-Au exhibited a relatively better discharge value compared to pristine graphene, particularly in the highest

resistance variation. Thus, the most suitable sample for use as an atmospheric ion harvester is the Graphene-Au sample, which can generate sufficiently high voltage and has a fast charging time as well as a relatively long discharge time.

The time constant values are obtained through exponential fitting modeling, and the choice of algorithm used can significantly impact these values. Some of the data show time constant values that do not align well with the voltage curve plot results in Figure 5. Therefore, the full charging and discharging times are also compared. The full charging time represents how quickly the data reaches its highest voltage during the charging phase. In contrast, the full discharging time reflects how long it takes for the data to reach zero voltage (the lowest point) during the discharging phase. When compared to the time constant values, the full-time values calculate the total time required for the charging/discharging process, while the time constant represents the time required for 63.2% of the entire process. The results of the full-time values calculation are shown in Figure 7.



**Figure 5.** Comparison of Full Time Values in Capacitor Charging and Discharging



In the calculation of full-time values for capacitor charging and discharging, it can be observed that graphene-Au tends to have a relatively fast charging time and a delayed discharging time in the data without resistance. Meanwhile, pristine graphene exhibits the fastest charging time and delayed discharging at full resistance. The results of

the full-time values calculation confirm that the addition of graphene layers to the copper electrode can improve the energy charging and discharging process times of the capacitor. This is particularly important in this study for storing energy harvested from atmospheric ions.

**Table 2.** Data of current, voltage, and power generated in the simulation

Electrodes	$I$ (mA)		$V$ (Volt)		$P$ (mW)	
	16 k $\Omega$	30 k $\Omega$	16 k $\Omega$	30 k $\Omega$	16 k $\Omega$	30 k $\Omega$
Pristine Graphene (Charging)	0.060	0.055	6.060	5.840	1.157	1.039
Graphene-Au (Charging)	0.108	0.057	4.580	5.830	1.376	1.026
Copper (Charging)	0.028	0.023	2.620	2.700	0.374	0.205
Pristine Graphene (Discharging)	0.109	0.051	6.060	5.840	0.796	0.354
Graphene-Au (Discharging)	0.121	0.080	4.580	5.830	0.664	0.559
Copper (Discharging)	0.099	0.044	2.620	2.700	0.311	0.144

Table 2 displays the current values, from which the power value is obtained by multiplying the current and the highest voltage value generated during the charging and discharging processes. Based on the data in the table, the energy produced by Graphene-Au is approximately 6.8 times and pristine graphene is 5.0 times greater than the energy that can be produced by copper alone. These results indicate that the addition of a graphene layer to the atmospheric ion harvesting electrode can optimize the process of generating electrical energy.

## CONCLUSION

In this study, the capacitive properties and ion harvesting characteristics of graphene-coated electrode samples were investigated. Raman spectroscopy results indicated that the graphene used in the study was a bilayer graphene. Meanwhile, CV tests showed a significant capacitance value for the graphene samples, specifically 0.40288 F for pristine graphene and 0.44879 F for graphene-Au. The addition of graphene was successfully proven to increase the energy output and slow down the capacitor discharge time in high-resistance ion harvesting processes through laboratory-scale simulation circuits. Based on these experiments, the energy generated by Graphene-Au was approximately 6.8 times greater, and pristine graphene was 5.0 times greater than the energy produced by copper alone.

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