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JEI Corrigendum

The original article was published Jun 30, 2020. On Sep 24, 2024, the authors noted that they introduced several errors during the preparation of the manuscript. The following corrections should be observed and are noted in red font:

1. The thickness of the graphene oxide structure is 0.34 nm (The paper only shows 0.34)

2. "PM1.0 describes particles with a diameter of 1 mm and smaller, PM2.5 describes particulates with a diameter of 2.5 mm and smaller, and PM10 describes particulates with a diameter of 10 mm and smaller." The definitions of PM 1.0, 2,5, and 10 are wrong. The diameter unit of the particle should be micrometers (μm), not mm.

3. The statement "However, the 2 mg graphene oxide per 100 mL ethanol group had the longest latency period before the PM2.5 concentration reached 250 g/m3, taking the longest time of about 500 seconds." g/m3 should be changed to μg/m3.

4. The statement "In addition, we calculated the breakthrough time, defined as the time it took to reach a PM2.5 concentration of 250/ for all the graphene oxide-coated groups. " unit of μg/m3 needs to be added.

5. In the summary part, "We observed that filters with the addition of graphene oxide were able to purify polluted air containing PM at concentrations above 3000\ \mu g/m3, a concentration above which commercially available filters cannot." The unit is mistyped. It should be μg/m3

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Article

Using graphene oxide to efficiently filter particulate matter at high concentrations

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SUMMARY

Air pollution is currently one of the biggest environmental challenges around the world. Air pollution not only causes detrimental effects to the human body but also endangers the natural environment. Many have proposed and created various methods to solve this problem, but none of them have worked very effectively. Following the discovery of graphene, scientists began to apply the use of this carbon material to address different kinds of problems. Using graphene oxide to combat air pollution, we prepared graphene oxide, which we then spread onto a commercial air filter, and used this air filter to purify air polluted with particulate matter (PM) of various diameters (PM_{10}, $PM_{2.5}$, or $PM_{1.0}$). We observed that filters with the addition of graphene oxide were able to purify polluted air containing PM at concentrations above 3000\ \mu g/m^3, a concentration above which commercially available filters cannot. The efficiency of graphene oxide-treated filters was much greater than commercially available filters. Based on our data, we proposed a mechanism by which graphene oxide can effectively diminish the amount of PM in the air.

INTRODUCTION

Graphene oxide is a two-dimensional single layer of graphite, a continuous carbon structure with a thickness of 0.34. Graphene oxide is arranged in hexagonal patterns with functional groups attached to the carbon structure (1). Graphene oxide has many unique physical and chemical properties, compared to other allotropes of carbon and metals. For example, graphene oxide has properties such as high conductivity, high transparency, and high carrier mobility (2). Additionally, because of its high surface area to volume ratio, graphene oxide is considered as a potentially useful material in many different fields. For example, graphene oxide can replace silicon in the manufacturing of semi-conductors, it can be used in making cell phone or solar cell batteries, and it is even being considered in the development of potential cancer therapies (2). Of interest to our group, we explored the application of graphene oxide in air purification. Specifically, we prepared highly purified graphene oxide and investigated its efficiency in purifying polluted air, comparing graphene-oxide treated filters to commercially available air filters.

Particular matter (PM) is defined as particles that float in the air. PM comprises most of the air pollution around the world and is separated into three categories according to particle diameter. $PM_{1.0}$ describes particles with a diameter of 1 mm and smaller, $PM_{2.5}$ describes particulates with a diameter of 2.5 mm and smaller, and PM_{10} describes particulates with a diameter of 10 mm and smaller. $PM_{2.5}$ is especially notable for its toxicity to the human body (3). In this paper, we showed that coating graphene oxide onto commercial filters increased the purification efficiency of air filters.

RESULTS

In this study, we investigated whether graphene oxide-treated air filters were more effective than commercially available filters in terms of purifying PM. First, using electrochemistry, we extracted and purified graphene oxide from used batteries. In order to confirm the identity and purity, we analyzed our graphene oxide sample using UV-Vis spectroscopy. We found that our extracted graphene oxide had almost the same peak absorption wavelength as previously reported for pure graphene oxide (**Figure 1**), confirming that our extracted graphene oxide had the expected properties of a relatively pure sample (4).

We first tested a set of controls: We used no filter, a commercial filter, and an activated carbon filter. We picked the commercial filter and the activated carbon since they are commercially available and commonly used. Notably, activat-

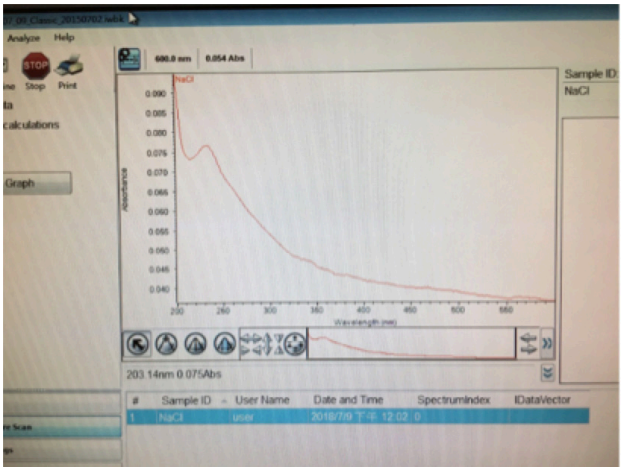

Figure 1. The UV-Vis spectrum of extracted graphene oxide sample. After extracting carbon rods from used batteries and purifying graphene oxide using electrochemistry, the purity of graphene oxide was confirmed using UV-Vis spectroscopy.

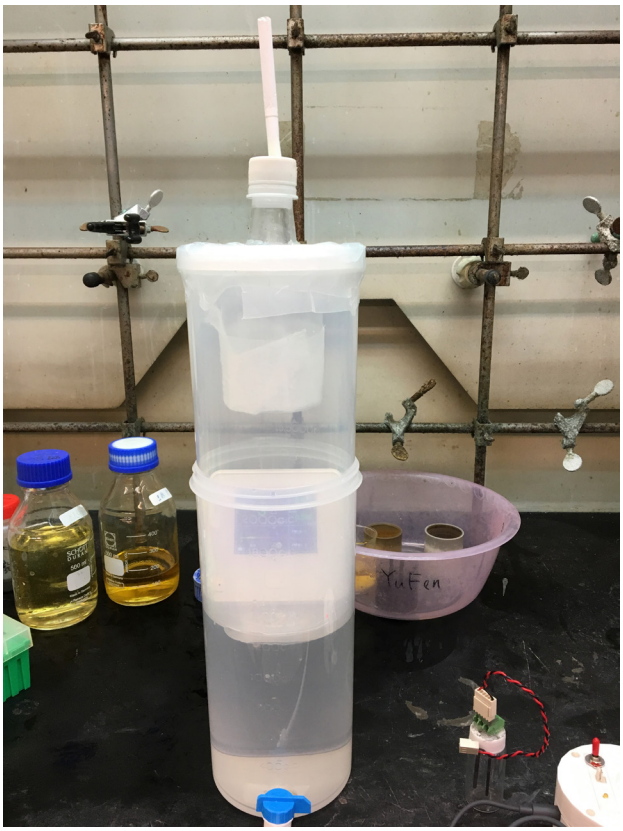

Figure 2. Experimental model. The setup was a simplified model of the human lung, consisting of several stacked chambers. When the cigarette at the top of the model was lit, air within the chamber was polluted with particulate matter (PM). As water at the bottom of the chamber was released, air was pulled from the top of the chamber, through the experimental filter, and reached the PM detector.

ed carbon and graphene oxide groups differ in particle size, since one particle of activated carbon is about 1000 times larger than that of graphene oxide. This leads to different filter spaces and may lead to differing performance. Of our experimental groups, we decided to test seven groups of commercial filters coated with differing concentrations of graphene oxide. We used 2 mg of graphene oxide dissolved in different volumes (50, 75, 100, 125, 150, 200 mL) of ethanol, to determine which group had the highest purification efficiency.

To test the purification efficiency of different filters, we built an experimental model in which we tested each filter's ability to purify PM from a cigarette (**Figure 2**). Our experimental setup was designed as a model of the lung. Water serves the function of a diaphragm in our model, because as the water flows out of the container (diaphragmatic contraction), the volume of the "empty" container (lung) becomes larger. As a result, air then flows into the chamber. In our experiment, we allowed water to flow out of the container after we lit a cigarette at the top of the experimental chamber. As the water flowed out of the chamber, air flowed through the cigarette smoke, pulling filtrate into the experimental filters. We measured the time it took for $PM_{2.5}$ within the model to reach 250 ug/m^3, the concentration considered to be hazardous to human life. The control, activated carbon, and untreated filter conditions took 50, 100, and 115 seconds, respectively, to reach 250 ug/m^3. In the graphene oxide-coated filter groups, it took about 90-280 seconds to reach this point (**Figure 3, Figure 4**)However, the 2 mg graphene oxide per 100 mL ethanol group had the longest latency period before the $PM_{2.5}$ concentration reached 250 g/m^3, taking the longest time of about 500 seconds.

Next, we compared the purification efficiency of filters treated with different graphene oxide concentrations, across different categories of PM. Specifically, we calculated both the purification efficiency and breakthrough time of the control and experimental groups (**Table 1**). In this experiment we concluded that the filters treated with 2 mg graphene oxide per 100 mL ethanol had the highest purification efficiency (**Figure 5**).

We also calculated the filter capacity, defined as the amount of PM held by a filter. We were interested in measuring how much more PM could be held by graphene-oxide coated filters than commercially available filters. We found that the filters treated with 2 mg graphene oxide per 100 mL ethanol had the biggest filter capacity of about 29 g/m^3 (**Figure 6**).

DISCUSSION

In our study, we hoped to create a filter that could address the problem of air pollution. We focused on removing PM, because PM is the main source of air pollution and has many detrimental effects on the environment. We aimed to capture an effective amount of PM in a given amount of time, supporting that removal of PM using our methods could become a good solution in tackling the current global problem of air pollution.

In our experiments, we coated air filters with different concentrations of graphene oxide in ethanol and tested their purification efficiencies. Across the different concentrations of graphene oxide, we found that all concentrations of treatment were able to purify PM more effectively than commercially available filters. Specifically, we discovered that filters treated with 2 mg graphene oxide per 100 mL ethanol had the longest

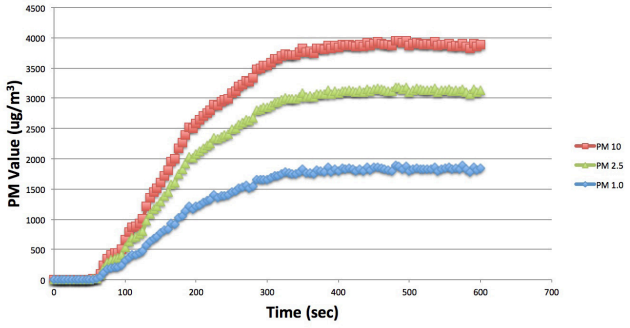

Figure 3. Concentrations of particulate matter over time following filtration using activated carbon filters. As a control experiment, an activated carbon filter was placed in the chamber under the cigarette. After lighting the cigarette and releasing water out of the chamber, allowing air to flow, the PM detector measured concentrations of PM with diameters of 1.0, 2.5, or 10 micrometers and less.

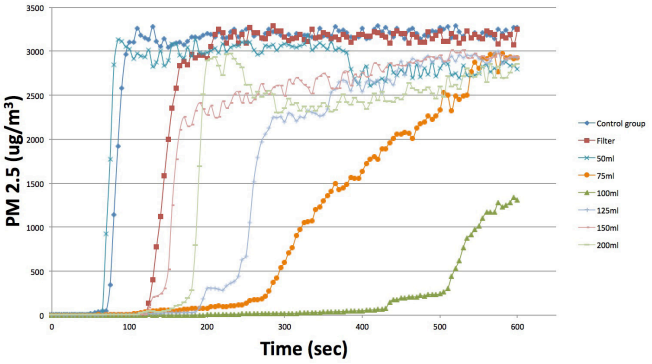

Figure 4. Concentrations of $PM_{2.5}$ over time following filtration using filters coated with different concentrations of graphene oxide. The PM detector was used to measure PM2.5 over time after the cigarette was lit and air was pulled through either no filter (control), a commercial air filter, or a filter coated with 2 mg of graphene oxide powder resuspended in various volumes of ethanol.

Purification Efficiency (%)	PM 1.0	PM 2.5	PM 10	PM 2.5 Breakthrough (250 ug/m3) Time
Control Group	N/A	N/A	N/A	50 Seconds
Commercial Filter (3M Filters)	95.56%	95.91%	95.75%	115 Seconds
Activated Carbon	97.55%	97.56%	97.57%	100 Seconds
2 mg Graphene Oxide/50 ml Ethanol	98.76%	98.83%	98.81%	90 Seconds
2 mg Graphene Oxide/ 75 ml Ethanol	99.88%	99.83%	99.83%	278 Seconds
2 mg Graphene Oxide/ 100 ml Ethanol	99.92%	99.96%	99.96%	502 Seconds
2 mg Graphene Oxide/ 125 ml Ethanol	99.82%	99.83%	99.8%	198 Seconds
2 mg Graphene Oxide/ 150 ml Ethanol	99.88%	99.83%	99.83%	141 Seconds
2 mg Graphene Oxide/ 200 ml Ethanol	99.76%	99.66%	99.65%	178 Seconds

Table 1. Purification efficiencies and breakthrough times of control and experimental groups. Values of purification efficiencies were reported for various tested filters across different categories of PM. Breakthrough time was calculated as the time required for the concentration of PM with a diameter of 2.5 micrometers or less to reach 250 ug/m^3.

breakthrough time, highest purification efficiency, and largest filter capacity of all other filters. Due to the surprising results of the 2 mg graphene oxide per 100 mL ethanol treatment, we decided to investigate the mechanisms that may have helped shape this purification effect. We came up with three possibilities to explain why the 2 mg graphene oxide per 100 mL ethanol group had the highest purification efficiency when compared to the other graphene oxide-coated filters.

First, the 2 mg graphene oxide per 100 mL ethanol group might have had the most even spread of graphene oxide over the filter. Secondly, graphene oxide contains several functional groups, such as hydroxyl and carboxyl groups, which could interact with polar PM molecules (1). Thirdly, the coated filter may have acted by using electrostatic adhesion to filter particles (1). However, more research should be done, perhaps through micro-structure analysis, to more fully investigate the reason why 2 mg graphene oxide per 100 mL ethanol had the highest efficiency.

In addition, we calculated the breakthrough time, defined as the time it took to reach a $PM_{2.5}$ concentration of of 250/ for all the graphene oxide-coated groups. We found that the 2 mg graphene oxide per 100 mL ethanol group had the longest breakthrough time, at about 500 seconds. Also, we calculated the filter capacity, defined as the amount of PM that graphene oxide-coated filters can hold, compared to commercially available filters. We found that the 2 mg graphene oxide per 100 mL ethanol group had the largest filter capacity of 29 g/m^3.

In our experiments, we concluded three main points to summarize the results of this experiment and highlight future directions. First, we successfully obtained graphene oxide powder using electrochemistry and measured purity with UV-Vis technology. Secondly, we determined that graphene oxide-modified filters had a higher purification efficiency, compared to a representative commercial filter. Moreover, the filter modified with 2 mg graphene oxide per 100 mL ethanol had the highest purification efficiency. In the future, we propose changing the organic solvent and experimental parameters to make further improvements on the purification process, and we will compare our filters to HEPA filters, filters that are designed by NASA to purify polluted air with an ability to filter 99.97% of the air particles with a diameter above 3. Additionally, in later experiments, we propose using micro-structure analysis to investigate the mechanism of air purification. Also, we will use organic solvents such as acetone because, while acetone serves similar functions as ethanol, it might produce different results since the chemical structure does not contain as many hydroxyl groups as that of ethanol (5). In addition, we will more definitively characterize the properties of our purified graphene oxide by analyzing any unexpected changes in carbon form or structure after being purified via electrochemistry.

Finally, we hope to find solutions that will help remove particulate matter from the filter, focusing on removing carbon

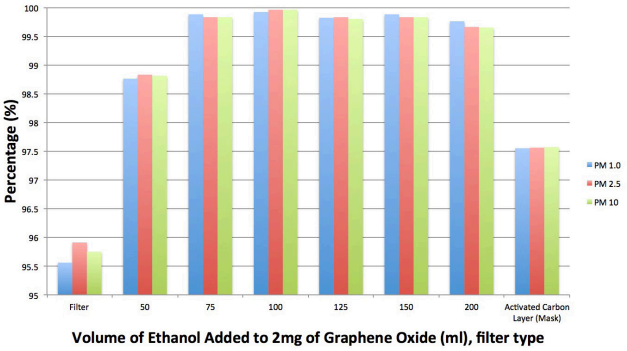

Figure 5. Purification efficiencies of different graphene oxide concentrations. The PM detector was used to measure concentrations of PM of different diameters. The filters tested were a commercial filter, an activated carbon filter, and filters coated with 2 mg of graphene oxide powder resuspended in various volumes of ethanol. Comparing the activity of each filter compared to no filter, purification efficiencies were calculated.

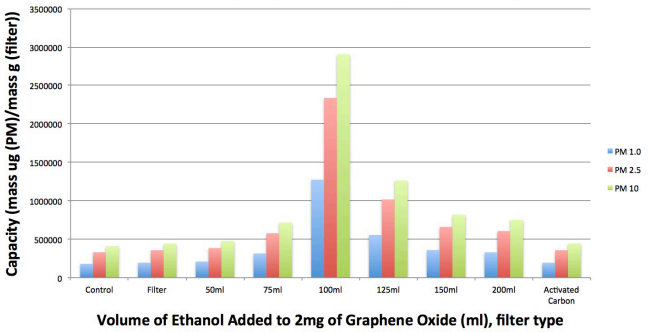

Figure 6. Filter capacities of different graphene oxide concentrations. The PM detector was used to measure concentrations of PM of different diameters. The filters tested were a commercial filter, an activated carbon filter, an uncoated filter, or a filter coated with 2 mg of graphene oxide powder resuspended in various volumes of ethanol. The filter capacities were calculated based on the saturated PM concentration of the control group.

monoxide since it takes up most of the chemical composition percentage in a cigarette, to make the filter reusable (6). This solution might help wash off PM on the filter and make the filter clean again. Because of this, a filter can be reused many times. With high purification efficiency and high reusability, coated filters are a potentially promising solution to addressing the world's air pollution problem.

MATERIALS AND METHODS

Purification of nano-graphene oxide

We used electrochemistry (4) to obtain highly purified nano-graphene oxide. We extracted carbon rods from used batteries (Panasonic NEO AA type battery), using 1 M sodium chloride as the aqueous solution and put two carbon rods in the solution. Next, we applied 10 Volts to the solution using a voltmeter for 1 hour. After electrolysis, we centrifuged the aqueous solution at 25000 RPM (rotations per minute) for 15 minutes, three times. After centrifugation, we incubated the solution for 1 day before testing its purity via UV-Vis spectroscopy. After testing its purity, the solution was lyophilized in the shelf freeze dryer to remove water for three to four days, resulting in with samples of graphene oxide power.

Applying graphene oxide to a filter

After extracting the graphene oxide powder, we mixed 2 mg of powder with different volumes (50, 75, 100, 125, 150, 200 mL) of ethanol by sonication, making sure that the powder was dispersed throughout the solution. Then, we put a 3M filter inside a petri dish and spread the graphene oxide/ethanol solution evenly onto the filter using a dropper. Next, we put the petri dish into an incubator at 60°C to separate the ethanol and graphene oxide.

Testing removal of particulate matter

At the top of our setup, we used a cigarette as the pollution source (Figure 2). Underneath that was our experimental filter to filter the polluted air. Below the filter was the PM detector, which can detect the concentrations of $PM_{1.0}$, $PM_{2.5}$, and PM_{10} of filtered air in order to measure the purification efficiency of each filter. At the bottom of the setup, we installed a water opening to let the water flow out of the container, in order to pull the air down so the polluted air could be filtered by the experimental filter at a steady rate.

After the experiment, we measured the efficiency of a filter by using the following equation:

$$Efficiency = 100 - \left(\frac{Particles\ allowed\ through\ filter}{Particles\ allowed\ through\ with\ no\ filter} \times 100\right)$$

Lastly, we also calculated the filter capacity of all the tested groups. In this case, we use the formula:

$$Filter\ Capacity\left(\frac{\text{mass(PM)}}{\text{mass(filter)}}\right) =$$

$$PM\ concentration\ rising\ time(seconds) * Flowrate\left(\frac{1000ml}{600s}\right) *$$

$$Saturated\ PM\ Concentration\ of\ control\ group\ (\mu g/m^3)$$

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