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Water Harvesting In Ambient Air

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Author
HUANG, XU

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Water Harvesting In Ambient Air

THESIS

submitted in partial satisfaction of the requirements for the degree of

MASTER OF SCIENCE

in Material Science and Engineering

by

Xu Huang

Thesis Committee:
Professor James Earthman, Chair
Associate Professor Yun Wang
Professor William Tang

2018
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ABSTRACT OF THE THESIS

Water Harvesting In Ambient Air

By

Xu Huang Anteater

Master of Science in Material Science and Engineering

University of California, Irvine, 2018
Professor James Earthman, Irvine, Chair

Even about 70% of our planet is covered by water; lack of access to clean water is still a pervasive problem disturbing people all over the world. Water problems are deemed to be worse in future, given the rapidly growing population. To solve these problems calls out for a tremendous amount of research to be conducted to develop new methods/technologies of economically purifying water and harvesting water, as well as reducing associated negative impact on the environment.

In this study, we investigated water condensation at a cooled surface and the impacts of the surface property and temperature. A device was designed and developed to collect the water condensation rate in a controlled environment. The Alumina surface was used as a substrate to collect water condensation under various cooling levels of the surface. Experiment data show that the water condensation rate rapidly decreases as the difference between the surface and environmental temperature goes down. Graphene is 2D carbon materials and it is a sheet of single-atom-layer thickness which consist of sp$^2$-hyridized carbon
atoms arranged in a hexagonal honeycomb structure. It offers many super
properties including high electrical and thermal conductivities, great mechanical
properties. To investigate its presence’s impacts on water condensation rate, the
Al surface randomly dispersed with graphenes was tested under the same
condition to study the impact of the graphene presence on the water
condensation rate. No obvious influence was identified. The study is important to
investigate the surface’s enhancement of phase change and water harvest from
ambient air.
1. Introduction

Cells that are the elements of human body need to be maintained with water, so water is the basic ingredient that supports all the organisms. No matter it is animals or plants, water supports the most basic life activities. Perhaps, people are so close to water that they are all habitual to ignore its importance. In addition, water is an indispensable material resource for the whole world, for example in agriculture people need to use water for irrigation, in animal husbandry people cannot separate from water in order to gain benefits. Thus our demand for water is tremendous. It seems that water is an inexhaustible resource on the earth because more than 70% of its area is covered with water, however, it is not the case. The water we mentioned above is usually fresh water which makes up only a small percentage of water on the earth.

<table>
<thead>
<tr>
<th>Source of water</th>
<th>Flow/km$^3$</th>
<th>Fresh water/%</th>
<th>Total/%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sea</td>
<td>1338000000</td>
<td>--</td>
<td>96.5</td>
</tr>
<tr>
<td>Glacier</td>
<td>24064000</td>
<td>68.7</td>
<td>1.74</td>
</tr>
<tr>
<td>Underground water</td>
<td>23400000</td>
<td>--</td>
<td>1.7</td>
</tr>
<tr>
<td>Fresh water</td>
<td>10530000</td>
<td>30.1</td>
<td>0.76</td>
</tr>
<tr>
<td>Salt water</td>
<td>12870000</td>
<td>--</td>
<td>0.94</td>
</tr>
<tr>
<td>Soil water content</td>
<td>16500</td>
<td>0.05</td>
<td>0.001</td>
</tr>
<tr>
<td>Underground ice</td>
<td>300000</td>
<td>0.86</td>
<td>0.022</td>
</tr>
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</table>

Table 1. Distribution of water resource on the earth
From table 1, we can know that fresh water makes up less than 3.5% of water resource on earth and most of it (about 68.7%) is the glacier, which means inaccessible. This can explain many problems worldwide related to the lack of clean, fresh water: 1.2 billion people can hardly get access to safe drinking water, more than 3900 children per day die from diseases caused by unsafe water, and 2.5 billion people have little or no sanitation[1]. And considering the rapid growth of global population drives up the demand in both developing and developed nations, the growth of water contamination, all these facts tell us that we need an economical and more effective method to reuse wastewater, without risking environment or human health.

Two-thirds of the world's population is dealing with water shortages[2]. It is ensured that the world's food needs are met by 2050 will take a doubling of global food production[3], which means the increase of the population will also at a rapid rate. If nothing is taken in future, no one can deny that the problem of water shortage will get worse. However, when you take a look at the composition and volume of air, you can know that the water vapor and droplets in the air are estimated to be about 13 thousand trillion liters[4]. This amount can support our global water consumption for more than three years. Also, this type of freshwater resource will be replenished gradually. Thus it is a key to solve our current water shortage problem.
2. Water harvesting

2.1 Introduction

In the natural world, all living creatures have their own way to acquire water. Some of them even possess a more effective way than human-beings.

Figure 1. Optical images of four typical creatures (spider silk, cactus, Namib desert beetle, and Nepenthes alata) and corresponding scanning electron microscope (SEM) images in nature, with the ability of water-harvesting[5]
Inspired by those excellent creatures, many researchers have done many types of research about water harvesting for decades and the main technologies are seawater desalination, wastewater reuse and water condensation.

2.2 Seawater desalination

Desalination is a process that extracts mineral components from saline water. More generally, desalination refers to the removal of salts and minerals from a target substance. Because it is a fantastic way to solve the freshwater shortage problem for those areas where near coasts. Thus for decades, many scientists have contributed their effort to get desalination accessible for commercial use.

Linares et al. studied forward osmosis (FO) hybrid systems’ applications in seawater desalination and wastewater reuse. Compared with conventional desalination technologies (such as reverse osmosis method), they have found that FO hybrid membrane systems possess a lot of advantages[6]: 1) It’s a more energy saving method. Through doing an equivalent work of desalination, forward osmosis system can save energy ranging from 72% to 85%. This is because forward osmosis requires no external hydraulic pressure during the treatment; 2) It owns long-term operation stability. For forward osmosis membranes, itself doesn’t induce suspended solids and other organic contaminants into the system[7]. Thus water flux decrease caused by fouling is much lower than reverse osmosis systems; 3) It is a more flexible treatment unit. Because it doesn’t need those external support makes forward osmosis systems
to have broader applications. Also, Linares has pointed out that there are still many challenges we need to overcome to get forward osmosis treatment to be applied in our daily life. The main problem is that, at this stage, we are lack of a high flux membrane which can ensure an elevated salt rejection, the availability of appropriate draw solution and a reduced internal concentration polarization effect.

Nanoporous graphene, which is expected to be an excellent material for desalination, has attracted many scientists’ attention due to the following merits: 1) Nanoporous graphene can have a high possibility to achieve 100% salt rejection because nanopores allow only ions and water molecules; 2) By the oxidative etching method and ion bombardment, high precisely sized nanopores can be acquired; 3) Various functional groups can be added to the graphene, thus, we can obtain a various desalination performance. However, Yi et al. pointed out that there are some drawbacks preventing nanoporous graphene from a broader application[8]. Firstly, it is really difficult to realize a high density of pores with a nano-size distribution, as well as water permeation will be restricted by nanoscale holes. Secondly, there is a limitation for obtaining continuous large-area perfect graphene and subsequent generation of nanopores when we scale up the production. Last but not least, those two technologies, ion bombardment and the oxidative etching technique, which are used to product nanoporous graphene are expensive methods. When implementing a scale-up production, the cost will increase dramatically. Therefore, Yi et al. presented us a
substitution. By controlling the pore size of graphene oxide membranes, it is still possible to exhibit 100% salt rejection in a cost-effective and simple way because both of raw materials and technologies are cheap[9]. In other words, this practical solution for filtration which can work as well as nanoporous graphene membranes for desalination with much lower cost.

2.3 Wastewater reuse

Wastewater is any water that has been affected by multiple human activities, including domestic, industrial, commercial or agricultural activities, surface runoff or stormwater, and any sewer inflow or sewer infiltration. Therefore, there are a lot of hazardous materials, such as various dyes, PPCPs and spilled oils, organics, and bacteria, in wastewater, all of them may jeopardize human health and environment[10-13]. To reuse wastewater, those hazardous materials should be extracted from wastewater.

Metal-organic frameworks(MOFs) have gained more and more attention due to their high porosity, facile modification and designable pore structures. All of these properties make metal-organic frameworks a good candidate for many areas, such as gas storage, adsorption, separation and delivery applications.

Inspired by MOFs, Zubair H et al. discussed the effect of MOFs on wastewater purification and analyzed plausible adsorption or interaction or interaction mechanisms behind it[14]. From the results, they realized that there are a
number of mechanisms, including electrostatic interactions, π-π stacking/interactions, hydrogen bonding, acid-base interactions, and hydrophobic interactions. In some cases, one mechanism will work alone but in other cases, multiple mechanisms might take place at the same time.

Figure 2. Possible mechanisms for how MOFs remove hazardous materials[14]

Organic dye pollutants, as one of main pollutants in wastewater, caused many problems when people try to purify wastewater. Graphene oxide(GO) sheets possess excellent adsorption capability for extracting organic dye pollutants from wastewater, which have attached more and more attention.

Guo H. et al. reported in his paper that they have prepared graphene oxide/polyethylenimine(PEI) hydrogen, working as efficient dye adsorbents, in a facile way. According to SEM data, between amine-rich PEI and graphene oxide sheets, the graphene oxide/PEI hydrogels were produced by both electrostatic interactions and hydrogen bonding. For two common organic dye pollutants,
methylene blue (MB) and rhodamine B (RhB), the GO/PEI hydrogels presented an efficient removal rate. They also posed that after absorption, the GO/PEI hydrogels can be separated from the wastewater. This means that the GO-based hydrogels for organic dye extraction and wastewater purification have a possibility for wide applications in future[15].

2.4 Water condensation

Water condensation is the process of water vapor turning into liquid drops of water. This process is very common in our daily life and it is the prerequisite for the formation of rain and snow.

Water condensation which occurs in ambient air can be described as that under the influence of high temperature, a parcel of air begins to rise and expand. However, as it going up, it will cool to the point where some of the water vapor molecules gather together and their thermal energy can never separate them again.

There is a heat cycle between water evaporation and condensation. At first, to evaporate, water will absorb heat from the surface of the Earth. It is critical for our environment because this process can maintain the Earth's surface climate much cooler which is necessary for reserving water otherwise there were no water on the Earth.
Then, during the process of water condensation, the heat extracted from the earth’s surface will be released and help the formation of clouds.

Except for occurring in ambient air, there is another way in which condensation occurs is on hard surfaces, for example during the formation of dew. There are some cases of this phenomena, including water condensing on the inner side of windows during winter season, or on the outer side of a ice coca, which are very usual. All of those examples are the result of those surfaces’ temperature are colder than the dewpoint of the air which is in contact with them.

Although desalination is a reliable way to get freshwater from seawater, it still has disadvantages preventing many countries from benefiting when people compare it with desalination. Therefore, water condensation has attained more and more attention due to it is more cost-effective and flexible to apply.

Anna Lee et al. utilized several parameters, such as the volume of drops that are collected from the surface, the speed of film condensation from unsaturated air and the growth rate of a drop via condensation, to investigate the effects of different surface wettability and subsequent water drainage on the rate of water harvest through vapor condensation.

For water condensation section, they found that a uniformly hydrophilic surface shows a higher water condensation rate than other two cases, a surface with hydrophilic patches in hydrophobic background and a uniformly hydrophobic
surface. This phenomena is in contrast to previous reports specific to fog basking. However, we can find that some animals in nature are using same strategy to obtain water, including Australian frogs and lizards.

For water drainage section, their tests shows a drainage path can provide an effective water exit as well as make up for the reduction of hydrophilic area. Nevertheless, only increasing the number of drainage paths for the current size of substrate does not change the performance of water condensation[16].

As mentioned earlier, metal-organic frameworks (MOFs), owning huge porosity, facile modification and design-able pore structures, are obtaining more and more attention in water condensation area.

Hyunho Kim et al. presented a design, which is based on a porous metal-organic framework {MOF-801, [Zr₆O₄(OH)₄(fumarate)₆]}, to collect atmospheric water. They also pointed out that there are several advantages for MOF-801 to become a potential material for water condensation: 1) An excellent water-adsorption performance. Many studies have proven that MOF-801 can have water-adsorption behavior on a molecular level; 2) Stability and recycling. After water-adsorption, MOF-801 can be separated and it is still stable; 3) Cost-effective. Because the constituent for synthesizing MOF-801 are very common and low-cost makes MOF-801 a cheap candidate.
This device, combined with a low-grade heat from normal sunlight, can collect water from air at ambient conditions, even when the relative humidity levels as low as 20%[17].
3. Applications of graphene in water harvesting

3.1 Introduction of graphene

Graphene is a material that has caught much attention in materials science, physics science and other many areas for sixty years[18-20].

Graphene is a crystalline allotrope of carbon with two-dimensional properties. It is a sheet of single-atom-layer thickness which consist of sp2-hydridized carbon atoms arranged in a hexagonal honeycomb structure[21]. This very special lattice structure[22-23] grants graphene to have many merits: 1) high pore volume; 2) large surface area; 3) high presence of surface functional groups in those materials.

Even though graphene is a two-dimensional material, its flexibility allows it to become a basic building units for all other dimensional graphitic materials. For example, 1) 0D buckyballs can be obtain by wrapping graphene up; 2) 1D nanotubes can be acquired by rolling graphene; 3) 3D graphite can be produced by stacking graphene.
3.2 Applications of graphene in water harvesting

There are multiple technologies that can be used for water harvesting, such as adsorption, flotation, disinfection, sedimentation, filtration, disinfection, membranes and desalination.

It has been mentioned earlier that graphene is very thin and it has really high porosity. All these advantages make many scientists to believe that graphene should have a good performance in water harvesting[24].

Figure 4. The transformation of how 2D graphene can turn into all other graphitic forms[24]
Cohen-Tanugi, D. and Grossman, J.C. used computational methods (CM) to present that nanoscale pores in single-layer graphene can filter NaCl salt from water in a very effective way [25].

Figure 5. The diagram of Nanoporous graphene membrane under working condition [25]

The performance of a desalination membrane depends on pore size, applied pressure and chemical functionalization. Their data shows that by controlling pore diameter of pores, the graphene membrane can effectively block the salt passage and allow for just water flow. Although the pressure values set for the computational experiments are distinctly higher than what is common for desalination in reality, the result that the time scales for flow scale is a linear function of applied pressure can support that their results will still work at a lower pressure. Besides, the hydroxyl functional groups in the graphene membrane can nearly double the water flux due to their hydrophilic character which the salt rejection performance declined as a sacrifice. However, the overall
permeability of water of this graphene membrane is significantly higher than conventional reverse osmosis membranes, usually several order of magnitude higher.
4. Experimental setup

In this study, we created a device to collect water vapor. The following figure is the device diagram.

Figure 6. Experimental setup for harvesting water through condensation

The main part of the device is a stainless steel bucket which is covered by a plastic plate. The bottom of bucket is filled with some water and because it is a enclosed environment, in very short time, the humility can reach 100%.

Due to the fact that water condensation is driven by temperature difference and water vapor will condense on a cooler surface. Thus, in this device, we applied a thermoelectric as cooling module to cool down the substrate (here is an aluminum plate) to create temperature difference. When we apply current on the thermoelectric, it will have different temperature on different sides. Because the
thermoelectric will produce high temperature on another side so a cooling system is placed on the thermoelectric’s hot side to extract heat away and ensure that it can work properly.

The whole process of water condensation in this experiment is that the water vapor will fill the bucket and when it gets close to the substrate whose temperature is lower than the inner environment it will condense on the substrate. After a period time, droplets will grow too big to be constrained by surface tension and then fall straight into a container.
5. Results and Discussion

5.1 Water condensation on aluminum plate

5.1.1 Simulation curve of water condensation

The condensation behaviors of the aluminum plate under different temperature difference can be measured by how much water a container can collect each hour. This measurement is also consistent with the equation of water mass transfer:

\[ m = D \frac{C_{T1} - C_{T2}}{R'} A \]  \hspace{1cm} (1)

Where \( D \) is the water vapor diffusion flux, \( C_{T1} \) is the concentration of water vapor at room temperature, \( C_{T2} \) is the concentration of water vapor on the cooler substrate, \( R' \) is a constant and \( A \) is the area of surface.

According to the equation of state of ideal gas, the concentration of water vapor can be expressed as:

\[ PV = nRT \]  \hspace{1cm} (2)

\[ C = \frac{P}{RT} \]  \hspace{1cm} (3)

Where \( P \) is the saturation pressure of water related to temperature, \( R \) is the gas constant and \( T \) is temperature.
5.1.2 Water condensation experiments

Since water condensation is a time-consuming process, we first investigated the water condensation performance for 9 hours to obtain measurable data.
Figure 7. The change of temperature difference under different condition during 9 hours

The figure shows the change of temperature difference for the 9 hours experiments. We have recorded temperature difference every ten minutes for the first and last two hours and every half hour for the middle five hours.

From the data, we can know that during first 2 hours the temperature difference for every experiment has presented a general tendency of decreasing, which means that the temperature difference was not stable during first 2 hours. However, for the rest 7 hours, the temperature difference only fluctuated a little bit.

The reason behind this phenomena is that, during the first two hours, water vapor will gather on the cooler aluminum plate and start to transform from gas phase to liquid phase. It is well-known that this process of water condensation is an exothermal process. Thus heat will be released during water condensation which in return will increase the temperature of the contact surface of aluminum plate and decrease the temperature difference between the aluminum plate and the ambient.
In order to acquire precise data, the average of the temperature difference of last 2 hours was taken as the final temperature difference for each experiment.

![Graph showing water condensation performance comparison between simulation and experimental curves. The simulation curve was created when the room temperature is 21.99°C (295.14K) and R’=40cm.](image)

From the above figure, we can know that both of two curves have shown a trend of increase when temperature difference goes up. However, the two curves are not perfectly overlapped. For every temperature difference, there is a gap between the simulation curve and the experimental curve.

It is very normal because the simulation curve is an idea situation but, in reality, there are many factors, such as dust and air agitation, that can have an influence on water vapor diffusion.

It has been mentioned earlier that, during 9 hours experiments, the temperature difference was not stable in the first 2 hours. This type of deviation can’t be
ignore because 2 hours’ uncertainty is too important to be neglected for a 9 hours experiment. Thus, the time period of every experiment has became 24 hours.

![Figure 9](image_url)

**Figure 9.** The comparison between results of 9 hours’ experiments and 24 hours’ experiments

There are two main reason for the change: 1) Compared to 9 hours experiments, 2 hours’ deviation can’t create much influence on the experiment anymore; 2) By elongating the time period of each experiment, a smaller temperature difference can be achieved.

A smaller temperature difference is indispensable for this experiment. The appearance of graphene on the aluminum plate will alter part of surface property of the plate and this change may have an influence on the performance of water condensation. When temperature difference is high the efficiency of water
condensation is dominate by the process of mass transfer. During this process, surface property can hardly affect it. However, the situation will switch to the opposite when temperature difference is low. Because under this condition, the effect of mass transfer is so limited and surface property of the substrate starts to dominant.

5.2 Water condensation on an aluminum plate with graphene

We used graphene dispersion as a graphene source and repeated all related experiments that have done before. By pressing the graphene dispersion under a certain weight, a graphene film has been attached on the aluminum plate.

In order to have a comprehensive observation of the effect of the appearance of graphene will have on water condensation, a waterproof HD camera has been set and taken a picture every 30 minutes.

(a) 00 mins (b)
Figure 10. The whole process of water condensation during first 4 hours. (a) is aluminum plate and (b) is aluminum plate with graphene.
From those pictures, we can realize that there is not an distinct difference between two cases. The process of water condensation occurred in two cases simultaneously. At 30 minutes, plates in two cases became wet. At 60 minutes, water vapor gathered on plate and started to form fog in both of two cases. At 90 minutes, it is obvious that water drops had formed. After 90 minutes, water drops grew gradually and they finally fell down under the influence of gravity.

Figure 11. The comparison of curves between 24 hours water condensation experiments with and without the presence of graphene

The comparison has shown that, with the appearance of graphene, the effectiveness of water condensation didn’t have a significant increase. When the temperature difference between 2-3°C, the graphene group seems to outrun the another group. However, it is not the case because there are some uncertainties in every experiment. Unless the difference is enormous to let us ignore those
uncertainties, otherwise, an affirmation that the presence of graphene will improve the performance of water condensation can’t be reached.

5.3 Sliding angle test

One important part for our experiments is to collect water. Basically, we can treat it as a wetting experiment so the sliding angle is a critical parameter for our experiments.

The sliding angle can be defined as a measure of the mobility of a droplet on a substrate[26]. According to Furmidge[27], the sliding angle is a function of the advancing and receding contact angles($\theta_A$ and $\theta_R$):

$$\sin(\alpha) = \gamma \frac{Rk}{mg} (\cos \theta_R - \cos \theta_A)$$  \hspace{1cm} (4)

Where $\alpha$ is the sliding angle;

$\gamma$ is surface tension;

$m$ is the mass of the droplet;

$R$ is the length scale constant for the contour of the droplet;

$k$ is the shape constant for the contour of the droplet.
Figure 12. (a) The force balance for a droplet inclined to the sliding angle. Those forces include the surface tension force ($\gamma$) and the body gravity force ($mg$). (b) Top view of the component of the surface tension force distribution that is parallel to the surface ($\gamma_\phi$) for a drop on an incline.

In this study, we also took pictures for both aluminum plates with and without graphene. The following pictures are the result of the shape of one small droplet at different angles.
Figure 13. The shape of a droplet at different angles with and without the presence of graphene. (a) is aluminum plate and (b) is aluminum plate with graphene.

By observing above pictures, it can be acknowledged that with the change of gradient, the shape of a droplet also changed. Besides, with the presence of graphene, the shape of the same droplet changed slightly. It turned from a more planar shape into a more round shape.

5.4 **SEM result of graphene**

Owing many excellent physical properties, graphene has become one of the most important materials among thousands areas. Therefore, we expected the presence of graphene could have a positive impact on water condensation. However, from all the results that we collected, the effect of graphene is not prominent. To explore the plausible reasons behind it, a graphene sample was characterized with SEM technique.
According to the SEM picture, the graphene flake can be observed clearly. Graphene still keeps its ordinary structure, however, the upper graphene sheet overlapped with lower one.

Basing on this fact, we analyse that there are two plausible reasons which could explain why the effect of graphene is so limited. The first reason is that the size of water molecule is too big to be impacted by graphene. Although graphene has many folds, the space between folds is too tiny for water molecule to enter. Thus the surface of graphene works as same as aluminum plate.

The another one is that the graphene sheets are overlapped makes the material on the aluminum plate more like graphite. Because the difference between
physical properties of graphene and graphite, the actual influence was caused by
the graphite which is so limited.
6. Reference


