Bio-Inspired Solar Electric Propulsion Investigation

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Abstract

Semiconductor materials frequently used in satellites and space solar panels are expensive and have risen technical and economic issues. Therefore, new alternatives to these solar panels are being pursued. Perovskite based solar cells have been of current interest in the past years due to their promising advantages in space technologies, such as their lightweight properties and their ease of manufacture. Yet, their sensitivity to moisture is a monumental problem that has prevented them from being utilized. In this report, we aim to induce hydrophobicity in these solar cells in order to address this problem. We used a multi-asset approach where we applied thermokinetics, molecular diffusion simulations, and data analytics**.** Also, various coating methods such as spin coating and drip coating to identify optimal perovskite deposition method and a combination of experimental methods including chemical and physical vapor deposition were used to add protective layers to the perovskite cells. We obtained a probable reaction mechanism between the perovskite material and water where the final products of the degradation were lead iodide (PbI_2) and aqueous CH_3NH_3I. This information was used to derive the rate law whose order of magnitude was found to be 2/3. We found that the optimal water repellant surface structure would be a combination of the convex epidermal texturing and the platelet epicuticular wax type. It was determined that dip coating is the most effective method of perovskite deposition and that initial chemical vapor deposition methods of salinization were not possible, while physical vapor deposition of Silicone Dioxide was feasible. Ultimately, these initial findings will provide insights into possible water resistant perovskite based solar cells.

Keywords: perovskite, epidermal, epicuticular wax, diffusion, hydrophobicity, solar electric propulsion

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I. Introduction

One of NASA Glenn Research Center Core competencies is Power, Energy and Storage Conversion, and further development of power technologies is required for deep space exploration. In order for mankind to reach the farthest reaches of the solar system, we must shorten the costs and weight required to develop spacecraft. Solar Electric Propulsion (SEP) is a project developed by NASA in order to develop spacecraft that will go further than any previous spacecraft. The main benefits of SEP is that it reduces the amount of propellant required tenfold by using electric power. NASA Glenn Research Center has been the innovator of power technologies in the past, and will spearhead this effort in developing revolutionary technology for space travel.[1]

We recommend the use of perovskite based solar cells for the development of SEP systems. Solar cells that are perovskite based have the potential to reduce the effect of arching, compared to other types of lightweight solar cells. Although perovskite solar cells are arching resistant, sustain high voltage, lightweight, they are subject to degradation in humid environments. Since there is no moisture in space, this would be the ideal scenario to utilize them, however, they must survive manufacture and launch on Earth in humid environments. Therefore, we aim to minimize the effects of moisture on these cells via hydrophobic surface level modification. We aim to extend the lifetime of the perovskites through modeling 1) the interactions between the perovskite and water, 2) the diffusion of moisture through a silicone layer, 3) data driven plant-based architectures. These models will inform experimental modification of the perovskite via the addition of encapsulants and hydrophobic layers. Ultimately, we aim to pass the NASA required AIAA-S111 standard qualification tests for photovoltaic arrays.

II. Methods

Through a variety of theoretical and experimental methods, we aimed to study, simulate, and test the interaction between water and the perovskite solar cells. We are able to modify the functional and protective layers of the perovskite solar cell in order to increase durability of these cells to external moisture. We combined the thermochemical modeling, theoretical visualizations, data analytics, and experimentation in order to understand the degradation of perovskite cells and maximize hydrophobicity.

A. Proof of Use

Currently, developing lighter solar arrays for space is a priority for NASA space missions, and perovskite based solar cells are not the only method of developing a light weight alternative to the traditional space-grade solar cells that NASA missions use. This is evidenced by recent NASA efforts to develop ROSA, or Roll-Out Solar Array. ROSA is lightweight and efficient, as are perovskite solar cells, however unlike perovskite solar cells they are already water resilient. In order to determine if further research in perovskite technology is warranted, a comparison between perovskite and another comparable technology, ROSA, was performed.

B. Thermochemical Models

The effect of moisture on a 15mm encapsulated thin fill perovskite solar cell was experimentally determined in a previous study by observing the diffusion of water through an epoxy layer.[2] The test cell was subjected to 20% RH for an extended period of time and the reduction of the perovskite material was measured in different time periods. Subsequently, by using the acquired experimental data, a generic moisture concentration profile in one dimension can be obtained by fitting the solution of Fick's first law of diffusion for our boundary conditions:

$$\frac{C}{C_o} = erfc\left(\frac{x}{2\sqrt{Dt}}\right) \tag{1}$$

Where C is the concentration of water, C_o is the concentration of water at the border of the cell, erfc is the complementary error function, x is the distance from the border of the cell, D is the diffusion constant and t is time. The diffusion constants at time t are then obtained by an extrapolating process. The desired profile at 1500 hours was calculated by using the equation of the linear regression of the relation between the obtained diffusion constants and time (Fig. 1):
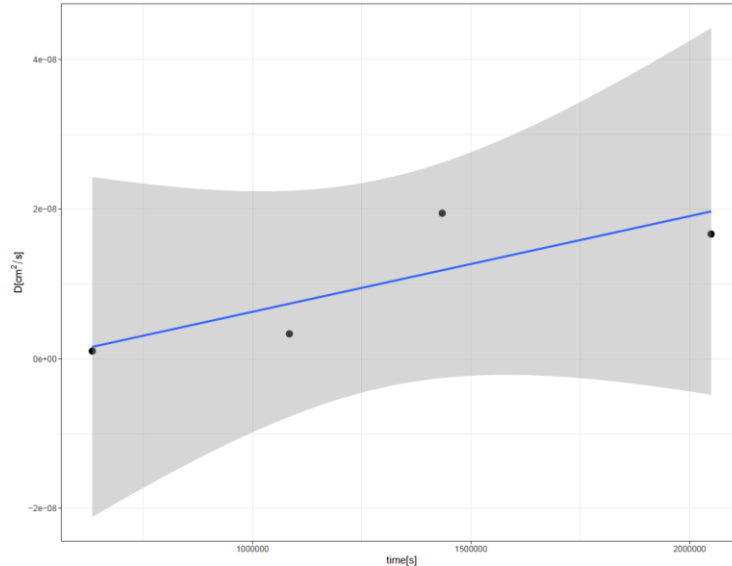
$$D = 2x10^{-7}t - 2x10^{-5} \tag{2}$$

Figure 1. Linear fit between the derived diffusion constant (D) at different time periods. The gray zone is the confidence level interval for predictions in the linear model.

The change in the Gibbs free energy of an open system due to a chemical reaction at constant temperature T and pressure P has to be adjusted by implementing the chemical potential μ of each species i as follows:

$$\Delta G(P,T) = \sum_i \upsilon_i \mu_i(P,T) \tag{3}$$

Where υ_i is the stoichiometric coefficient of the reacting species i. For an ideal gas behavior, the chemical potential can be defined as:

$$\mu_i = \mu_i^\circ + RTln(P_i/P_\circ) \tag{4}$$

Therefore, taking the ideal gas law we can relate the pressure of species i in terms of concentration $C = P/RT$. Taking this assumption and equation x, the change in Gibbs free energy can be estimated for various RH conditions as follows:

$$\Delta G_{rxn} = \sum \mu_i^\circ + RTln\left(C_s erfc\left(\frac{x}{\sqrt{2Dt}}RT\right)\right) \tag{5}$$

C. Diffusion Simulation

Diffusion is the motion of one or multiple particles due to a concentration gradient. That is, the particles move from a state of high concentration to a state of low concentration. The motion of this particles is random and is referred to as Brownian motion. In Physics, the random walk algorithm is used as a simplified model to explain molecular Brownian motion and diffusion. In a random walk all the particles have the equal probability of moving in any direction. Using a diffusion simulation can enable us to understand the behavior of water molecules on the surface of the silicone layer, and how the water will degrade the perovskite solar cell. As water molecules keep adding up onto each other, they will form droplets, which will eventually surpass the silicone layer and degrade the solar cell (perovskite). The more the solar cell degrades the less efficient it becomes.

The simulation was developed using Java high level programming language. The particles were modeled using a hard sphere approach. Hard spheres are defined as impenetrable objects that don't overlap in space. They resemble the strong elastic repulsion that atoms and spherical molecules undergo at very close distances. The particles are under constant velocity and their mass is not taken into account as our priority was to simulate the random walk.

The particles were animated using the Java.awt package which is used for GUI and animation development. The particles start in the left wall of the simulation since the only exposed areas on which water could enter the silicone encapsulant is through the corners. We also chose the left corner as the standard entry point for default. The particles start diffusing over the surface and start moving further from the starting point. A sticking layer was added on too. The particles stick for a certain period of time on the sticking layer and a sticking coefficient determines the time they will stick for. If the sticking coefficient is 1 for example, then the particles will stick for 1 second.

The simulation changes the number of particles every minute, so on the first minute the number of particles is 100, on the second minute the number of particles is 200, on the third minute it is 500, and on the fourth minute the number of particles is 1000. When the timer resets to 0, the number of particles starts again at 100, and the sticking coefficient increases by one. This process goes on and on until it reaches a sticking coefficient of 5.

D. Data Acquisition and Modeling

Through data acquisition of hydrophobic plant structures using AskNature and scholarly journals, variables that affect hydrophobicity were defined and organized to be used in the data modeling for optimal hydrophobic surfaces. Hydrophobicity means the water will minimize its contact on the surface and form compact liquid droplets because of the tendency water molecules have to attract one-another referred to as cohesion. The data collected by Barthlott and Neinhuis[3] provided a lot of information for the surface structures of super-hydrophobic plants including contact angles. A contact angle is the angle the water droplet forms when it touches a surface and it determines hydrophobicity. When the contact angle is less than 90 degrees the solid surface is not hydrophobic. If it is larger than 90 degrees it is considered hydrophobic. When the angle is larger than 150 degrees, the solid surface is considered super-hydrophobic and has high self-cleaning properties figures 2-5.

Super-hydrophobic surfaces in plants show a combination of epidermal micro-structure texturing and epicuticular wax nano-strucutures which in turn show high water repellency. Water droplet on these hierarchical structures may touch on the top of the nano-structures without fully sitting on the surface because of air pockets filling in the gaps of the structures underneath them. As described by the Cassie-Baxter model, this dual roughness will affect the contact angle.[4]

$$\cos(\theta_c) = f_s(1+\cos(\theta_\gamma))-1 \qquad (6)$$

According to the Cassie-Baxter state, the θ_c or the contact angle that determines hydrophobicity increases as the fraction of the solid surface in contact with the liquid or the f_s decreases. The θ_γ refers to the ideal contact angle in an ideal solid flat surfaces and even if it is less than 90 degrees, the actual contact angle θ_c can still be increased. So, as the droplet minimizes its contact on the surface, it forms compact liquid droplet as a cause of cohesive forces pulling inward. These droplets will then roll off the leaf surface collecting any dust particles in its way.[5] During data mining, variables like contact angle, epicuticular wax type, epidermal texturing and whether the plants maintained water repellency throughout its lifetime were collected.
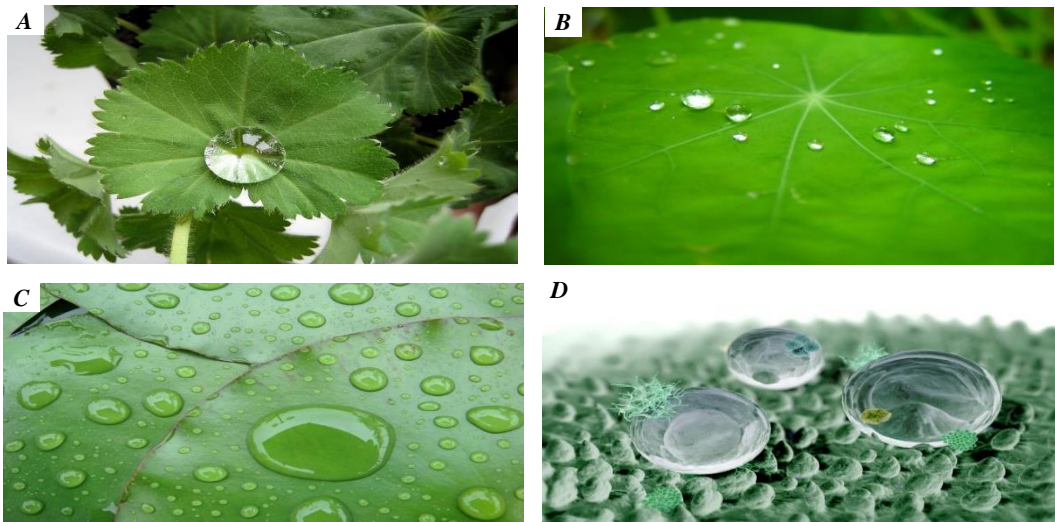

Figure 2. Images of hydrophobic plants and their rough surface structures. The water droplets stay spherical in their surface layer because of the cohesive forces. They will eventually roll off carrying the dirt with them and cleaning the leaves. Figure 2D shows the surface structures and water droplets in micro-scale.

E. Experimental Fabrication

Perovskite fabrication methods were optimized through experimental methods. Non-functional perovskite solar cells were developed in order to study the effect of various superficial preparation methods on the resistance to humidity of the perovskite materials. Additionally, the thickness of the perovskite layer was measured as a means of

optimizing the darkness of the solar cell functional layer. Traditional experimental thin film coating methodology was used to coat the samples, and space-grade encapsulant was used to protect the cells.

The addition of a silane Self Assembled Monolayer, or SAM, through chemical vapor deposition on the cells was tested as a potential means of achieving chemical hydrophobicity. Previous research conducted on CIGS (Copper indium gallium selenide) photovoltaic cells, which like perovskites are sensitive to moisture, proved that the addition of a silane layer would protect the cells form degradation. As seen in Figure 3, the addition of an APTES ((3-Aminopropyl) triethoxysilane) substantially minimized degradation in efficiency of these after hours in damp heat. We hypothesize that the same effect could be had on perovskite solar cells. We hypothesize that the free hydroxyl groups that react with water and cause degradation could be inactivated through a salinization process, chemically halting the cell degradation. APTES, as seen in Figure 4, has three free ethyl groups that could theoretically bond to the hydroxyl groups of concern.
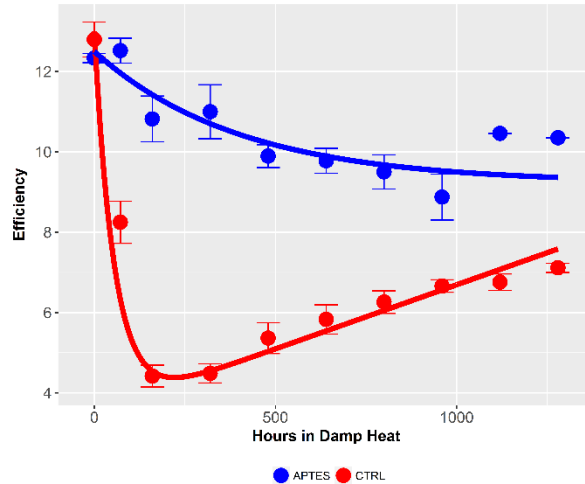

Figure 3. Degradation of the efficiency of CIGS (Copper indium gallium selenide) in damp heat environment with and without a protective APTES ((3-Aminopropyl)triethoxysilane) layer. While CIGS saw some recovery over time due to degradation, the silane protected cells exceeded the control in maintenance of their efficiency.[2]
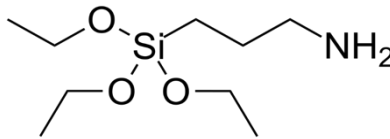

Figure 4. Structural Formula of APTES.

For the salinization process, a thermal Chemical Vapor Deposition (CVD) process using APTES dissolved in toluene was used. Toluene was chosen as the solvent as it is used in the fabrication process of certain types of perovskite cells. The solutions were developed outside of the glovebox in an air environment then the CVD process took place outside of the glovebox in a nitrogen environment.

Another addition of a Silicon Dioxide (SiO_2) as an experimental dielectric surface passivation layer was conducted to ensure that the oxide would not degrade the perovskite. The SiO_2 was deposited through Physical Vapor Deposition using the Angstrom Deposition Chamber. While this layer may not have an effect on general hydrophobicity, it may be necessary to add other functional layers of the perovskite cell later in the fabrication process, and so it was of interest to us.

Ongoing steps of this project include placing the experimentally developed cells in an environmental test chamber and testing for degradation via light transmission measurements.

III.   Results and Discussion

Every part of the project had different results to consider. First a visualization of the diffusion of water was created and the diffusion of moisture through encapsulation was also investigated to understand how water particles diffuse on surfaces and degradation occurs. Through experimentation, we discovered more about the perovskites advantages and issues. By collecting data of super-hydrophobic plant surfaces, the optimal water repellent surface structure based on plant surfaces was found. Together these results offer different ways to tackle the issue of moisture on the perovskites and their degradation.

A. Comparable Light Weight Solar Cell Efforts

While we know that perovskite solar cells have the potential to be beneficial, we conducted a theoretical study to determine if they would be competitive with other impending technology that NASA is researching. Based on this theoretical comparison between the two, ROSA has a higher energy output per surface area unit, given generous efficiency assumptions (ROSA at 30% and perovskite at 20%). This result, however, proves that perovskite solar cells are on par with other modern technologies, and substantially exceed the requirements of energy output per surface area that has been sufficient for space travel in the past. The ISS, with a solar panel area of 2500 m[6], has an energy output of only 120 kWh[6] Because ROSA is significantly subject to arcing and would be much more difficult to manufacture on a large scale, it was concluded that continuing research in a perovskite alternative concurrently would be beneficial to accomplish current NASA missions.
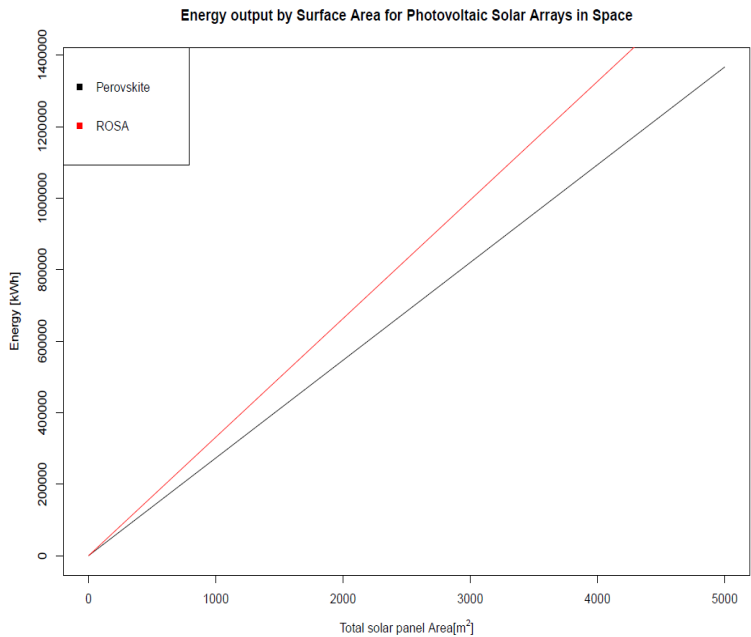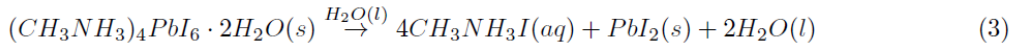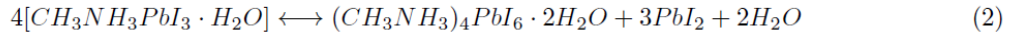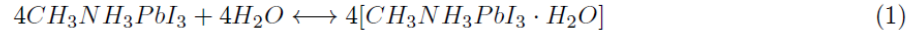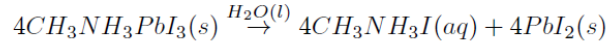

Figure 5. Comparison of the relationship between energy output and solar area in traditional ROSA (Roll-Out Solar Array) and thin-film perovskite solar cells. These thin films are competitive with other modern high-efficiency solar cells, while more cost efficient.

B. Possible Degradation Chemical Path and Rate Law

It has been shown that the degradation mechanism of perovskite films under humidity depends strongly on various parameters; such as the humidity percentage, temperature, and light conditions. For example, the degradation process can be reversible under 35% RH.[7]However, for the purposes of this study, we aim to obtain insights of the behavior of these cells when they are exposed to 95% RH and at 45 °C. Leguy et al. degradation path gives an appropriate hypothetical pathway for the degradation of $CH_3NH_3PbI_3$ due to moisture:

$$4CH_3NH_3PbI_3 + 4H_2O \longleftrightarrow 4[CH_3NH_3PbI_3 \cdot H_2O] \qquad (1)$$

$$4[CH_3NH_3PbI_3 \cdot H_2O] \longleftrightarrow (CH_3NH_3)_4PbI_6 \cdot 2H_2O + 3PbI_2 + 2H_2O \qquad (2)$$

$$(CH_3NH_3)_4PbI_6 \cdot 2H_2O(s) \xrightarrow{H_2O(l)} 4CH_3NH_3I(aq) + PbI_2(s) + 2H_2O(l) \qquad (3)$$

overall reaction

$$4CH_3NH_3PbI_3(s) \xrightarrow{H_2O(l)} 4CH_3NH_3I(aq) + 4PbI_2(s)$$

The first step in the mechanism, is the formation of a monohydrate phase when the crystal structure is saturated with one water molecule, this is followed by the formation of a dihydrate phase upon exposure longer to moisture accompanied by the formation of lead iodide (PbI_2). When step 2 is completed and the entire structure is in excess water, $CH_3NH_3^+$ dissociates and irreversibly degrades the cell. This mechanism under the desired conditions (45 °C, 95 % RH, in the dark) is strongly hypothetical since the effect of the temperature was not taken into account in the experiments of this study. Also, since the intermediate phases were not obvious in some XRD experiments, it can be concluded that step 3 is the rate-limiting step. With the proposed mechanism followed by a slow step, the rate of the degradation reaction can be derived. The suggested rate law is $r = k[CH_3NH_3PbI_3]^{2/3}$, with an overall order of 2/3 where $k = k_3(108K_2K_1[H_2O])^{2/3}$. The constants K_2 and K_1 are the equilibrium constants of step 2 and step 1, respectively. And the concentration of water, $[H_2O]$, can be calculated by using Fick's first law:

$$[H_2O] = C_s erfc\left(\frac{x}{2\sqrt{Dt}}\right) \qquad (7)$$

This rate law suggests that the degradation rate is faster at higher RH.

C. Diffusion of Moisture through Encapsulation

By reason of the protective glass layers at the top and bottom of the cell, once the cell is exposed to humidity, condensed liquid water diffuses through the sides as seen in Figure 6. Water diffuses through the material and the degradation of the perovskite begins; this process increases with exposure to higher humid environments. Figure 7 shows a generic representation of the experimental 15 mm test cell exposed to humidity for an extended period of time. Where x characterizes how far the degradation has deteriorated the cell through time. In order to know how much material should enclose the cell to stop the degradation process, Fick's third law was used as means obtain possible x values. Results from the previous experimental
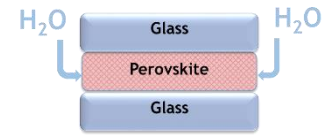

Figure 6. Representation of a perovskite based solar cell exposed to water molecules.
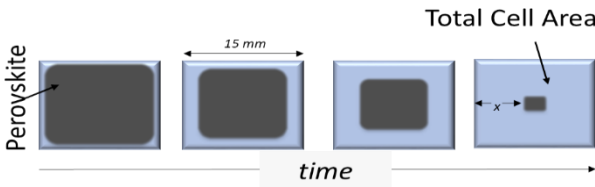
study are shown in Figure 8, where the concentration of condensed liquid water at the surface of the encapsulant over the length of the cell is displayed for different exposure times to humidity. As can be seen, the photovoltaic cell degradation is significantly higher when exposed to higher time periods. For our test requirements (95 % RH and 1500 hours) it can be inferred that the length of material needed is approximately 0.2 cm per 15 cm.

Figure 7. Degradation of an encapsulated thin fill perovskite solar cell in time.

Therefore, the actual perovskite structure should be inserted after this length in order to reduce the deterioration process caused by water.
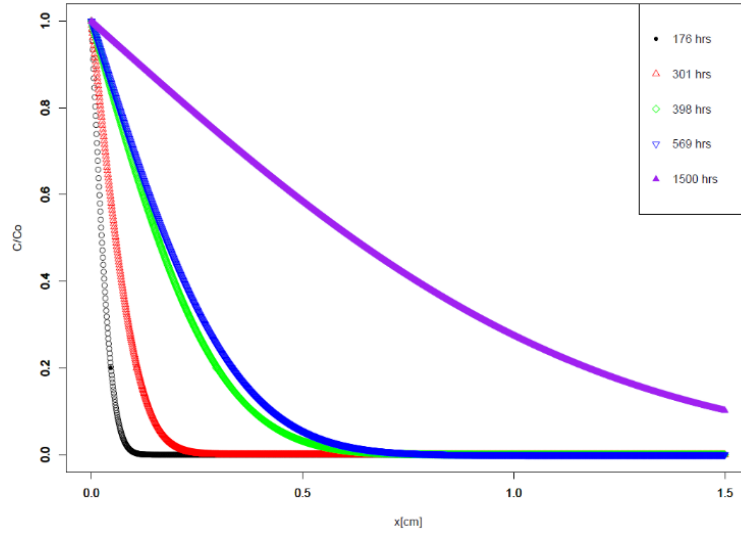
Figure 8. Degradation of Solar Cells due to Moisture. The effect of the relative humidity (RH) in the degradation of thin-film Perovskite solar cells for various time periods.
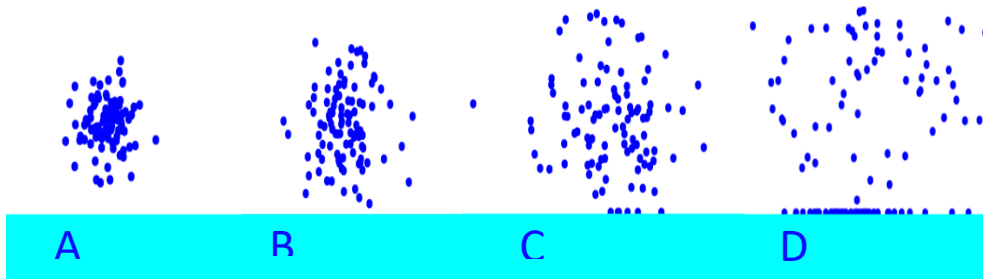
D. Diffusion Simulation

Figure 9. Diffusion simulation particles start from the center.

Figure 9 shows a diffusion simulation of water particles where the particles start in the center of a simulation box (Fig. 9A). As the simulation progresses over time, the particles move farther away (Fig. 9B and Fig. 9C) until they reach equilibrium (Fig. 9D) where the particles stay more or less on the same space. The surface layer has a sticking coefficient of 2 and therefore, the particles stick to the surface for 2 seconds until they are set free.
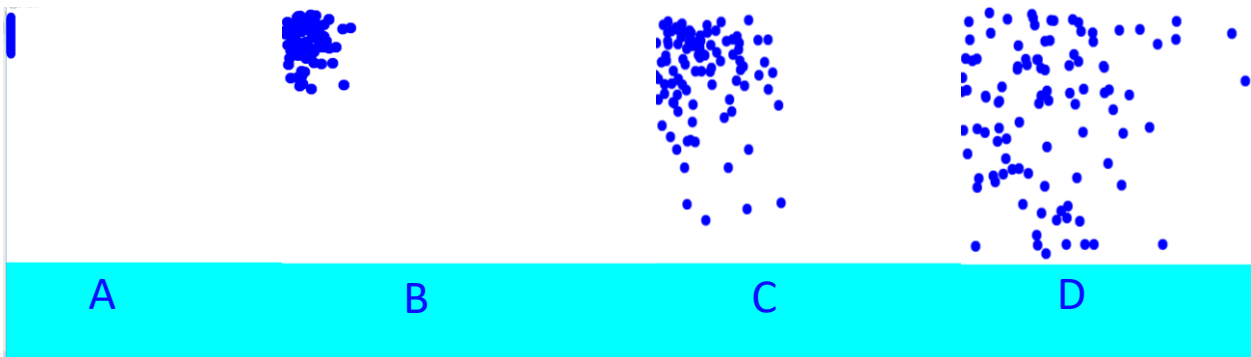

Figure 10. Diffusion simulation particles start from the upper left corner.

Figure 10 shows the diffusion simulation where the particles start on the left corner (Fig. 10A) as time passes they move farther to the right (Fig. 10B and Fig. 10C), although the particles still tend to be on the right (Fig. 10D). The particles stick to the surface for N seconds, depending on the sticking coefficient, since it can vary, the bigger the sticking coefficient the more time the particles will stick to the surface layer, also the number of particles varies overtime.
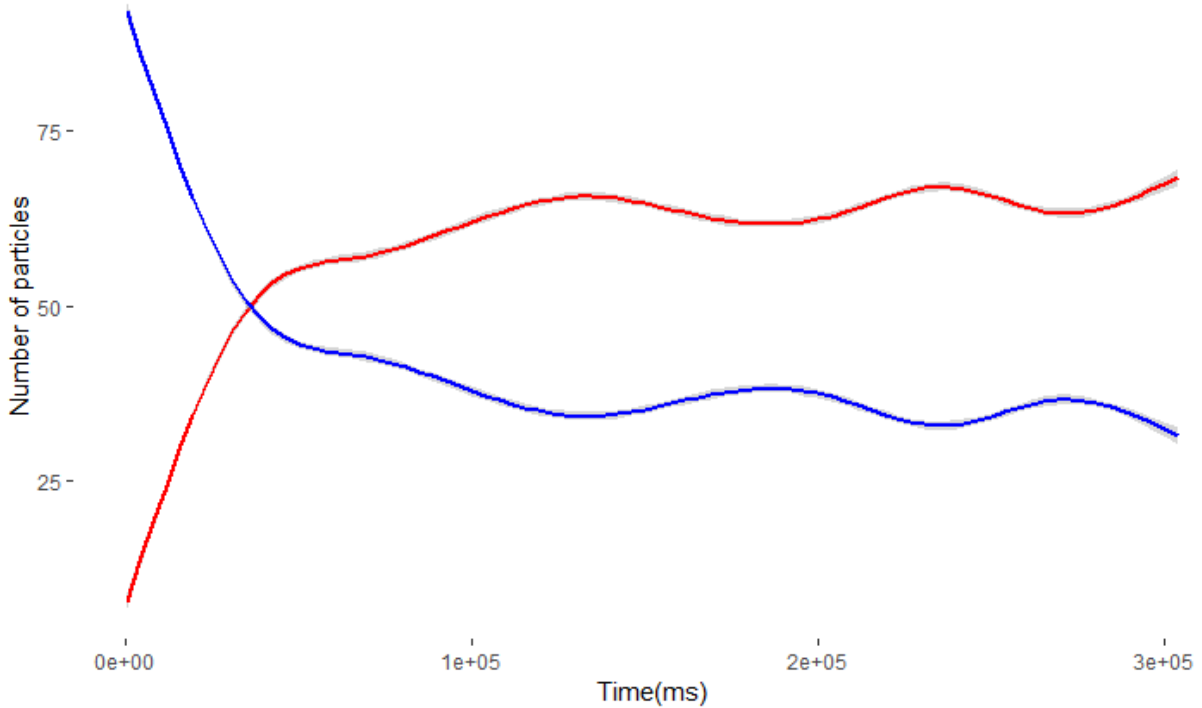

Figure 11. Graph depicts stuck particles vs free particles for a sticking coefficient of 1.

Figure 11 shows how initially all the particles are free, but as time goes on they start sticking to the surface, we can see there is a point on which we have equal number of stuck and free particles. As we reach this equivalence point the particles enter a state of equilibrium, on which the number of free particles and stuck particles will remain constant.

E. Optimal Hydrophobic Surface Structure

By modeling the data, we can make predictions on the optimal hydrophobic structures. Boxplots were utilized to show the contact angle of super-hydrophobic plants containing a combination of either convex or papillose epidermal cell structure with either epicuticular wax structures: platelets, rodlets or tubules. The LMWR refers to the lifetime maintenance of water repellency, where the red boxes represent the plant surfaces that did not maintain hydrophobicity throughout its lifetime, and the blue boxes did. The type of epicuticuclar wax structure with the papillose structure appears to have no effect on the contact angle, as opposed to the convex structure which contact angles varies by the epicuticular wax structures. There might be a hidden variable that needs to be considered for the papillose-epicuticular wax types combinations. The optimal super-hydrophobic surface structure is determined to be the combination of convex and platelets as it can be seen in Figure 12.

There were difficulties in this study like a lack of information and data for the hydrophobicity of plants. Most of the data available was for super-hydrophobic plants only (contact angle is more than 150 degrees), and certain epidermal and wax structures were considered. However, a mathematical model could be drawn in future research that will include more diverse structure types.
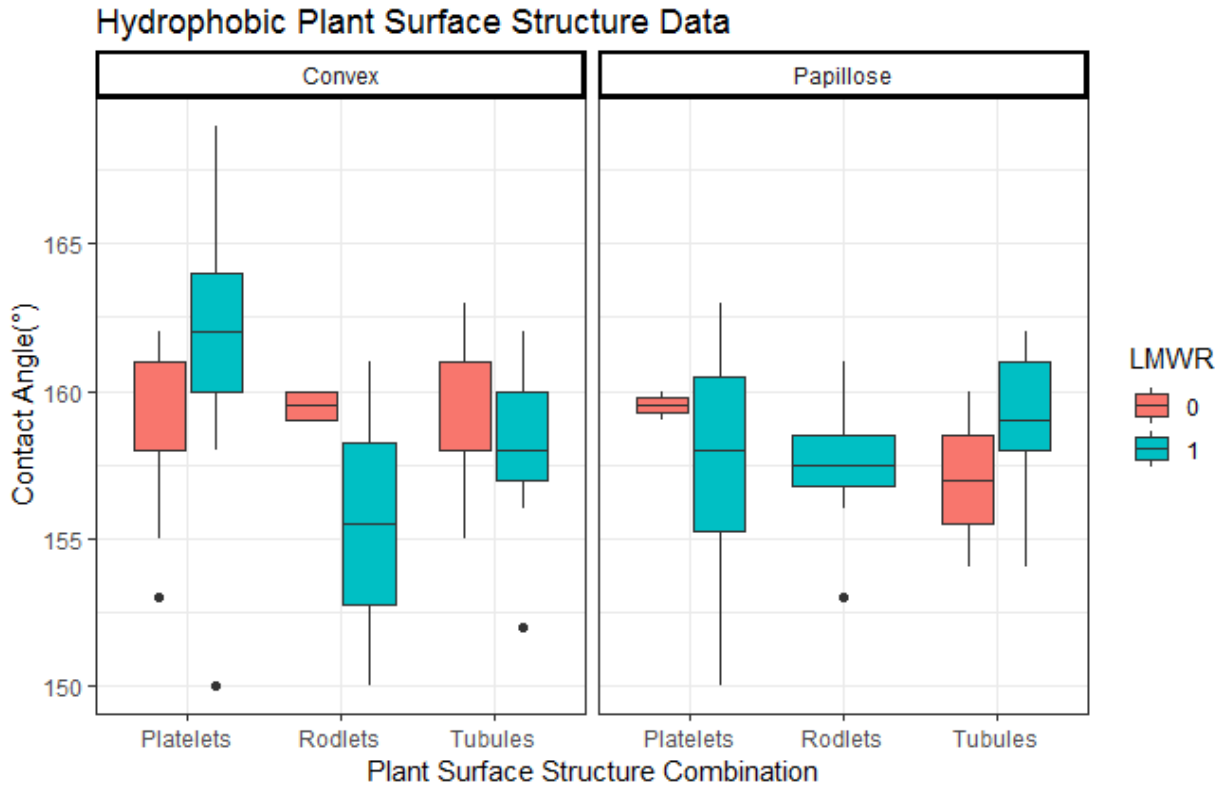
Figure 12. Graph showing the contact angle of super-hydrophobic plants containing a combination of either convex or papillose epidermal cell structure with either epicuticular wax structures: platelets, rodlets or tubules. The LMWR refers to the lifetime maintenance of water repellency. The type of epicuticuclar wax structure with the papillose structure appears to have no effect on the contact angle, as opposed to the convex structure which data spread is affected by the epicuticular wax structures.

F. Experimental Fabrication and Protection

The aforementioned silanizaiton process was proven to be ineffective. After a standard CVD process of 1% APTES dissolved in a toluene solution, the perovskite cells were fully degraded. While it is unclear as to why this occurred, some theories include:

1) That the APTES itself degraded the perovskite simply by bonding to the free hydroxyl on the surface of the perovskite
2) That the toluene used had dissolved H_2O in it, which reacted with the free hydroxyl on the surface of the perovskite, degrading it
3) That the toluene itself reacted negatively with the perovskite, causing degradation

In order to test the first theory, we suggest future research to spin coat perovskite samples pure APTES. While it would likely not result in a SAM the way that CVD could, this would prove if the APTES itself has properties that degrade the perovskite. In order to test the second and third theories, an anhydrous toluene kept in a nitrogen glove box could be used, because it would not have dissolved water in it and therefore if the perovskite degrades through CVD with a pure Anhydrous Toluene solution, this was likely the reason for degradation before.

While the Chemical Vapor Deposition process with the prior protocol proved to be ineffective, the addition of a SiO_2 layer of 100-150 nm did not degrade the perovskite. This proves that an oxide layer would not degrade the free hydroxyl on the surface of the perovskite. This is promising for the addition of a hydrophobic SAM, despite the degridaiton of the perovskite due to the CVD process described prior.

Through initial experimental fabrication, it was quickly determined that the manufacturer described methods of perovskite precursor deposition and silicone encapsulation were not viable. Perovskite deposition via dynamic spin coating at 4000 rpm yielded layers that were too thin and transparent, while they were meant to be dark charcoal and opaque. A variety of deposition methods were tested, from testing the spin coating at various speeds, testing dynamic

vs. static deposition methods, and drip/dip coating methods. In order to maximize the coating, it was determined that a drip method would be most yield thickest results, yet least uniform. Additionally, and spin coating at speeds lower than 1000 rpm yielded uneven results but thicker layers. Through this, it was determined that the ideal deposition method for the future could be inkjet printing, as it would allow for thicker deposition while maintaining uniform layers. As for the silicone encapsulation, the manufacturer suggested ratio of 10:1 silicone to curing agent was not able to cure in nitrogen in the 24 hours advertised. Rather, a ratio of 8:1 cured on a heat above 100°C was most effective. It is unclear as to where these inconsistencies occurred, however mitigating them is critical to developing perovskite test cells.

Once the amended protocol has been finalized, future directions include placing the perovskite test cells in an Environmental Test Chamber at 95% humidity, and testing the change in transmittance of light over time. Because as perovskites degrade they change from a dark charcoal color to a clear yellow color, transmittance is an effective method of measuring their degradation. Through these measurements, it is possible to determine which perovskite cells have the best initial performance as well as which are most sensitive to humidity and degradation over time.

IV. Conclusion

By using computational and theoretical modeling, data acquisition and experimental methods, we got insights into possible water resistant perovskite based solar cells. Our ultimate goal is to improve these solar cells' durability and implement them as a means for solar electric propulsion in space missions. We will advance photovoltaic power technology by providing cheaper and lighter weight alternatives. This technology would enable us to accomplish NASA missions in a more cost effective way by for example, enabling us to make frequent trips from the ISS to the Moon. As concluding remarks, this project has the potential to revolutionize the method through which solar energy is harvested for propulsion in space travel and will allow us to push boundaries and overcome seemingly insurmountable obstacles, such as interplanetary travel.

Acknowledgments

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