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## Studying Phase And Morphology Of Hydrocarbon Petrochemicals In Inorganic And Organic Microdroplets Using An Electrodynamic Balance

Fafa Tackie-Otoo

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STUDYING PHASE AND MORPHOLOGY OF HYDROCARBON  
PETROCHEMICALS IN INORGANIC AND ORGANIC MICRODROPLETS  
USING AN ELECTRODYNAMIC BALANCE

by

Fafa Tackie-Otoo

Bachelor of Science Mechanical Engineering, Kwame Nkrumah University of  
Science and Technology, 2016

A Thesis

Submitted to the Graduate Faculty

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In partial fulfillment of the requirements

for the degree of

Master of Science

Mechanical Engineering

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Organic Microdroplets Using an Electrodynamic Balance

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To my ever-supportive husband, Nii Otinkoran and son-to-be Nii Okai.

## ABSTRACT

Wastewater from the petroleum industry contains large amounts of inorganic salts, hydrocarbons, mercaptans, oil and grease, phenols, and other organic chemicals. All these compounds can be found in highly complex solutions in the petroleum industry's discharged water, and there is no common treatment method, which precludes reuse paths in other sectors such as irrigation of farmland. In order to reuse produced water, the water must be treated and these harmful chemicals must be removed. To treat and potentially reuse oily waste outside the energy sector, researchers typically use membrane technology, photocatalytic degradation, oxidation processes, electrochemical catalysis, and other methods. The goal of this project is to introduce a novel method to measure chemical thermodynamic properties, such as water activities, of produced water. I demonstrate that the electrodynamic balance (EDB) technology is beneficial for exploring properties of produced water that can be applied to treatment and reuse in the future.

## NOMENCLATURE

AS	Ammonium sulfate
AA	Atmospheric aerosol
CA	Citric acid
DF	Damping factor
DRH	Deliquescence relative humidity
DBET	Dual balance electrodynamic trap
EDB	Electrodynamic balance
ERH	Efflorescence relative humidity
FT	Free troposphere
RH	Relative humidity
GA	Glutaric acid
GF	Hygroscopic growth factor, $D/D_0$
HTDMA	Hygroscopicity tandem differential mobility analyzer
IPCC	Intergovernmental Panel on Climate Change
$M(\text{dry})$	Dry mass of the particle
mfs	Mass fraction of solute
NaCl	Sodium chloride
Nd:YAG	Neodymium-doped yttrium aluminium garnet
PW	Produced water
Q	Flow rate
$V(\text{bal})$	Voltage balance
$\mu\text{m}$	Micrometer
nm	Nanometer
Sccm	Standard cubic centimeters per minute
TDS	Total dissolved salts

## 1.0 INTRODUCTION

### 1.1 Relationship between produced water (PW) and atmospheric aerosol (AA)

In this chapter, I introduce the sources and importance of produced water (PW), atmospheric aerosol (AA), and I also explain the relationship between them. First, PW is generated by the oil and gas energy sector; it is the fresh water that emerges from the well along with the crude oil during extraction. As a result, PW contains suspended solids, dissolved solids, soluble and non-soluble oil/organics, and other chemicals utilized in the production process.<sup>1,2</sup>

Over the course of the well's life, the proportion of produced water to oil changes. This ratio typically exceeds three and, in some regions of the world, can even exceed twenty.<sup>3</sup> The composition of the produced water changes with time in addition to the flow rate. From well to well, generated water has a vast range in chemical make-up.

Similarly, AA are also highly variable in their chemical composition due to complex atmospheric chemical processes that govern particle formation and growth, and individual particle properties. Similar chemical compounds are present in PW and AA, owing to the overall abundance of these compounds and their inevitable mass exchange between bulk water sources and the atmosphere, such as by evaporation of volatile and semi-volatile petrochemicals.

I draw this similarity because understanding PW can shed light on AA, and vice versa; furthermore, I explore the possibility of applying techniques that have advanced the study of AA in recent years to be of potential benefit to understanding PW. Because of the large amounts of PW generated annually by the energy sector in the United States, it is necessary to explore options for treating it for potential uses in other sectors that benefit society using scientific techniques that are typically applied to studying AA.

Despite the widespread belief that oil and water do not mix well, separating the two usually necessitates several processes. The level of produced water treatment is determined by the site's treatment needs, which are often surface water discharge, deep well injection, reinjection, or evaporation ponds.<sup>4</sup> Beneficial reuse is becoming a more attractive choice as rules have tightened, disposal methods have gotten more expensive, and water is becoming scarcer. Treatment of produced water is frequently regarded as an upstream oil and gas process. Removing oil and suspended particles from water can be difficult depending on the degree of chemical and mechanical emulsification.

As of now, produced water cannot be used outside the energy sector as it can pose significant challenges including high expense of treatment, the resulting water's potential for longterm toxicity, and public acceptance.<sup>5</sup> Additionally, a "one size fits all" solution is unlikely because the quantity and characteristics of the produced water change over time. The goal is to improve treatment technology by first understanding the solutions to be treated.<sup>6</sup>

## **1.2 Using Aerosol Instrumentation to Study Produced Water Microdroplets**

Numerous instruments can be used to detect how aerosol particles react to variations in RH. It has been shown that the electrodynamic balance (EDB)-based particle levitation approach is an effective way to examine the hygroscopic characteristics of individual aerosol particles.<sup>7</sup> This method has the benefit of enabling continuous monitoring of the particle mass as a function of RH, allowing for in situ unambiguous characterization of the particle mass growth due to water uptake. Particles with sizes greater than a few micrometers can use it. The hygroscopicity tandem differential mobility analyzer (HTDMA) is an alternative technique for measuring water uptake.<sup>8</sup>

In this work, I harness EDB technology to investigate using a Dual Balance Electrodynamic Trap (DBET) to measure phase transitions of briney aqueous droplets relevant to

the energy sector. The key advantage of studying produced water in microdroplet form is that EDB instruments and other microparticle levitation platforms access extremely high solute concentrations known to be found in produced water.<sup>9</sup> However, the estimates of total dissolved salts (TDS) in produced water are close to supersaturation with respect to solute, which creates difficulty in using conventional methods to study produced water at these metastable concentrations. I start my tests using only binary solutions containing water and NaCl, due to the well understood chemical thermodynamics of aqueous NaCl in both bulk and droplet form, as well as the fact that NaCl is the most abundant salt in the Bakken oil and gas play in North Dakota.<sup>5</sup> This project lays a foundation for near future work to accomplish exploring salt mixtures and other salt compounds, such as bromides and sulfates, as well as petrochemicals such as organic acids.

Although this thesis focuses on improving wastewater reuse in the energy sector, the mixtures I examine are also atmospherically relevant. The chemicals found in produced water are volatile or semi-volatile, and therefore partition to the atmosphere and ultimately contribute to aerosol particle loading. Therefore, it is pertinent to understand the environmental problem of how energy sector wastewater impacts atmospheric aerosol. Next, I will explain the background of how atmospheric aerosols affect the environment.

### **1.3 Particulate Matter (PM)**

In the atmosphere, aerosol particulates are referred to collectively as particulate matter (PM), which includes mineral dust, raindrops, fog, smog, smoke, soot, and haze.<sup>10</sup> Depending on the origins, aerosol particles can be categorized as follows: direct emissions of primary particles from human or biological sources, secondary sources are created by chemical processes in the atmosphere. The primary sources of atmospheric aerosols can be divided into the following groups: (i) common surface sources of primary aerosols, such as arid soils (mineral dust), the ocean



(sea salt), the biosphere (pollen), biomass (mostly OC), and fuel combustion (OC and EC); (ii) diffuse sources, such as air traffic, cloud evaporation, secondary aerosols produced by the chemical synthesis of volatile substances (organics), and extraterrestrial sources, within the atmospheric volume (vaporizing meteors), and (iii) powerful point sources, such as a volcano.<sup>11</sup>

The majority of the world's aerosol emissions come from natural sources, but in urban areas, the relative anthropogenic share increases significantly. Aerosol particles have a lifespan that can run from a few hours to several days, the latter long enough to permit long-distance transport. The primary characteristic of aerosols is their variability and heterogeneity in time and space as a result of their multiple sources and comparatively brief lifetime when compared to several trace gases. Wet deposition is the primary drain. The coarse mode has significant sinks for both dry and moist deposition. Their lifetime in the stratosphere may be similar to the number of years an air parcel spends there, allowing for worldwide distribution and transit.<sup>10,11</sup> The earth's radiation equilibrium is impacted by aerosol particles in the atmosphere in a variety of ways.<sup>12</sup>

Aerosol particles, first and foremost, absorb and disperse radiation. The hygroscopicity of the aerosol particles, which is primarily decided by their chemical composition, has an impact on this direct aerosol effect. Second, both the chemical composition and the size distribution of the aerosol particle are related to the propensity for cloud formation and the resulting cloud properties.<sup>13</sup> The indirect aerosol effect is the result of this impact on cloud albedo and the radiative characteristics of cloud droplets.

Inorganic and organic components can be distinguished among atmospheric aerosols. Most inorganic salts found in the atmosphere have well-known hygroscopic characteristics.<sup>14,15</sup> The hygroscopic characteristics of quite a few pure substances have been studied among the numerous organic species found in the aerosol.<sup>16</sup> However, only a small number of mixtures' hygroscopic

characteristics have been studied thus far. Inorganic salts, such as ammonium sulfate (AS) and sodium chloride, can exhibit hysteresis behavior during the uptake and loss of water, i.e., by displaying a difference between the deliquescence and efflorescence relative humidities (DRH/ERH) and with a higher water content of the deliquesced than the effloresced particles in this relative humidity (RH) range. In contrast, organic aerosol components without hysteresis behavior can help the aerosol absorb water at a lower RH than the DRH of inorganic ions. This has been documented by <sup>17</sup> and is also theoretically anticipated given that such complex mixtures can maintain a liquid state and exchange water with the gas phase at lower RH's.<sup>18</sup>

Organic aerosols, which are primarily produced by the photochemical oxidation of primary organic matter and volatile organic compounds (VOCs), are a significant component of fine particulate matter (PM). These aerosols have a detrimental effect on human health, air quality, and climate change.<sup>19</sup>

#### **1.4 Organic aerosols**

A significant but extremely variable portion of atmospheric aerosol is made up of organic compounds. Direct emissions of organic aerosol particulates occur from both biological sources; vegetation and the ocean, and human sources; the burning of fossil fuels, biomass, and industrial processes.<sup>20</sup> Along with these particle emissions, there is also a gas phase emission, and the oxidation-based degradation of these precursors results in the creation of secondary organic aerosol (SOA). The anthropogenic effect is apparent in the major contribution, i.e., biomass burning. The main component of biomass-burning particles is organics.<sup>21</sup> Depending on the region, aerosol particle chemical composition varies greatly.

In the haze plume from the United States, measurements over the Atlantic showed that organic aerosols dispersed at least as much light as sulfate particles. There are indications that

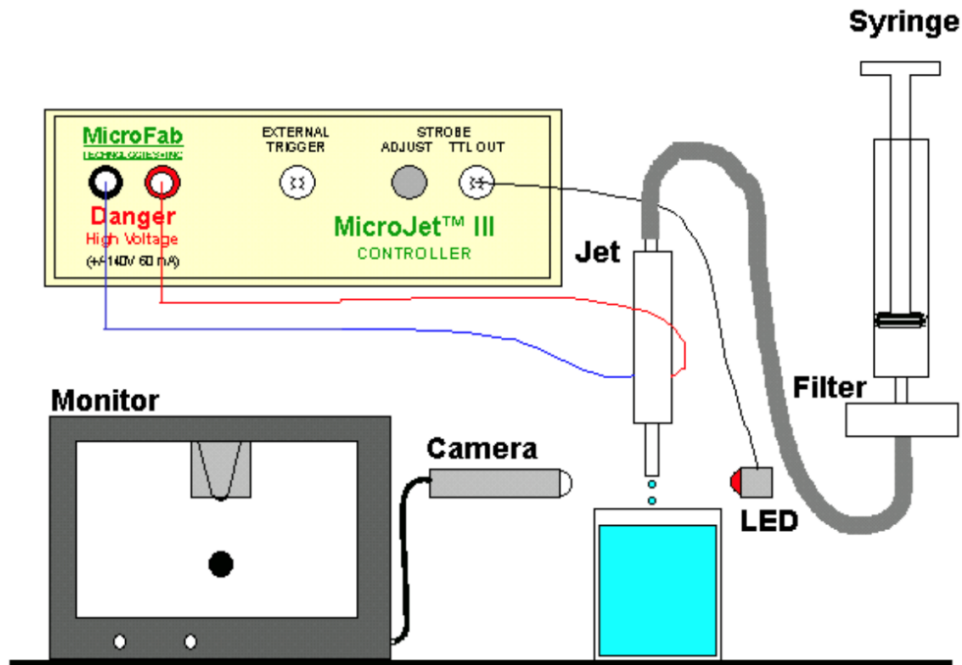
organics occur in the lower stratosphere and are also significant components, if not the majority, of upper-tropospheric aerosols.<sup>22</sup> Many of the organic molecules in aerosols are water soluble due to the presence of polar functional groups, especially carboxylic and dicarboxylic acids. This influences the aerosol particles' water uptake and enables them to engage in cloud droplet nucleation<sup>23</sup>, and<sup>24</sup> Recent field measurements have shown that organic aerosols may function as effective cloud nuclei and, as a result, have a significant impact on the indirect climate influence.<sup>25</sup>

## 2.0 EXPERIMENTAL METHODOLOGY

### 2.1 Advantages of Using Inkjet Dispensers

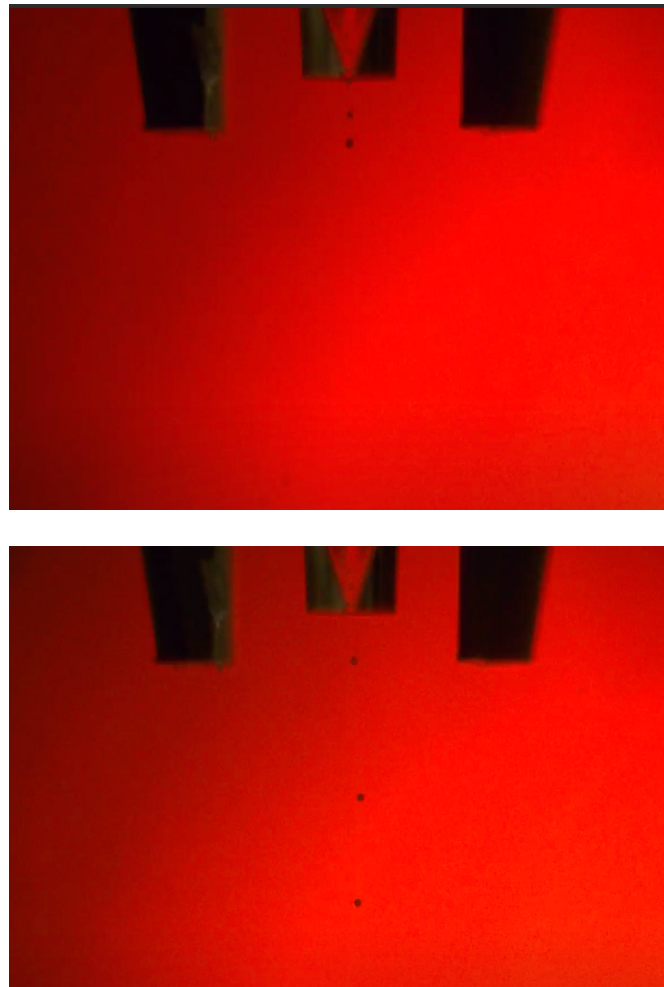
Inkjet dispensers are typically used in microparticle levitation experiments due to their reliably controlled sizes of the droplets that are generated and their droplet-on-demand capability.<sup>29–32</sup> Setting up operational parameters is the simplest configuration for an inkjet dispenser. It needs a controller to create the pulses, a dispenser with a reservoir and a bucket to dispense into, and an observing system (Figure 1).

An easy-to-use syringe can supply the fluid and back pressure (Some pressure control function, which for the present purpose is fairly basic, is provided by the combination of the piston of the syringe and the operator's thumb). A fixed reservoir, such as a closed vial or bottle, and a pressure system to deliver controlled back pressure to the fluid reservoir through a designated intake in the cap should be used instead of a syringe for a more serious procedure.



**Figure 1:** Basic jetting setup. A second driver, not depicted in the figure, is typically used to connect the strobe LED that illuminates the droplets for the camera, MicroFab Technologies, Inc., User's Guide, June 2012, Version 4.1.

A "Pneumatics Console" from MicroFab Technologies provides respectable manual pressure control. An Alicat controller, for which support is included into the JetServer™ application, can also be ordered. A different range of pressure control is required in some applications, such as the dispensing of fluids with high melting points; MicroFab provides a Tescom ER3000 pressure controller for this use, which is operated via a different computer interface on the same control computer. The camera will observe an almost motionless shadow of any droplet in its field of view when the strobe LED illuminating it is flashed with very brief pulses (usually approximately 10 $\mu$ s duration); see Figure 2.



**Figure 2:** *Drop formation with Sodium Chloride (NaCl).*

## 2.2 Product Description

My project requires dispensing microdroplets directly into an electrodynamic trap. Therefore it is worthwhile to explain the product's use in detail. MicroFab's drop-on-demand single jet dispensing devices of the MJ-A design are ideal for dispensing droplets of aqueous and solvent-based fluids at varied temperatures. Because of an integrated orifice and virtually entirely glass wetted surfaces, MJ-A devices are particularly resistant to hostile fluids. I can improve their process by selecting drop volumes within a two order of magnitude range by using orifice sizes ranging from 20 to 80  $\mu\text{m}$ . Fluids with viscosities less than 20 cPs and surface tensions ranging from 20 to 70 dynes/cm can be dispensed.

The integrated orifice extends from the housing, providing visibility of both internal flow and drop creation. Metal extensions on both sides can optionally protect the orifice from injury. Microdrop dispenser heads work on the same idea as an inkjet printer with a piezoelectric print head. A capillary tube is used to feed the liquid. A piezo ceramic imparts a mechanical impulse to the liquid column to emit a droplet. A microfine droplet is formed at the nozzle tip and exits the dispenser head at high speed. The size of the drop (15-120  $\mu\text{m}$ ) is determined by the capillary opening and driving mode. The electronic control of the dispenser heads ensures a high level of dispensing precision in terms of volume, timing, and positioning. The operation is visible thanks to an embedded stroboscopic LED.



MJ-ABP-01

**Figure 3:** *Dispenser Head.*

### **2.2.1 Drop-on-Demand Mode (DOD)**

In drop-on-demand mode, the hydrostatic pressure created by a fluid level higher than the orifice and backpressure (low vacuum) are balanced to keep the fluid meniscus at the dispenser orifice. Pressure from the piezoelectric actuator is employed to form droplets in this case. The dispensing program controls the ejection of one droplet every trigger of the device. Because the reservoir is above the orifice level, a tiny vacuum is required in drop on demand mode to prevent dripping at the orifice.

To dispense, input a waveform parameter in the software that will eject the droplet. Each fluid has a unique set of waveform parameters that are determined through an iterative process. As a starting point, create a unipolar waveform with a rise and fall time of  $3.0\mu\text{s}$ , a dwell period of  $30\mu\text{s}$ , a voltage of 30V, and a frequency of 240Hz.

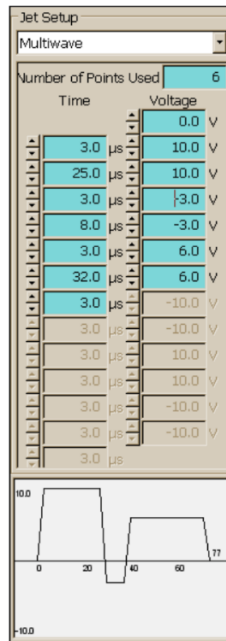
Experimentation is used to investigate the interaction between the dispensing and catch fluids when collecting microspheres. Drop on demand microsphere dispensing is commonly accomplished by holding the dispenser tip in air over the catch solution, but I do not require this in this particular project.

### **2.2.2 Finding useful operating parameters**

The proper operation of a jet, and its verification, is typically the first step in my experiments to ensure that the microdroplet size is in my desired range. It must be large enough to be visible in the trap. Within the range of 20 – 50 micrometers is ideal for my experiments. The following is the standard process:

- Position the jet over a waste bucket, such that the horizontal camera can see the tip of the device and some 2 mm below it.
- Fill the jetting device.

- Ensure that the tip of the device is free of pending droplets.
- Make the device run in continuous mode and adjust pulse shape parameters until good jetting is observed.



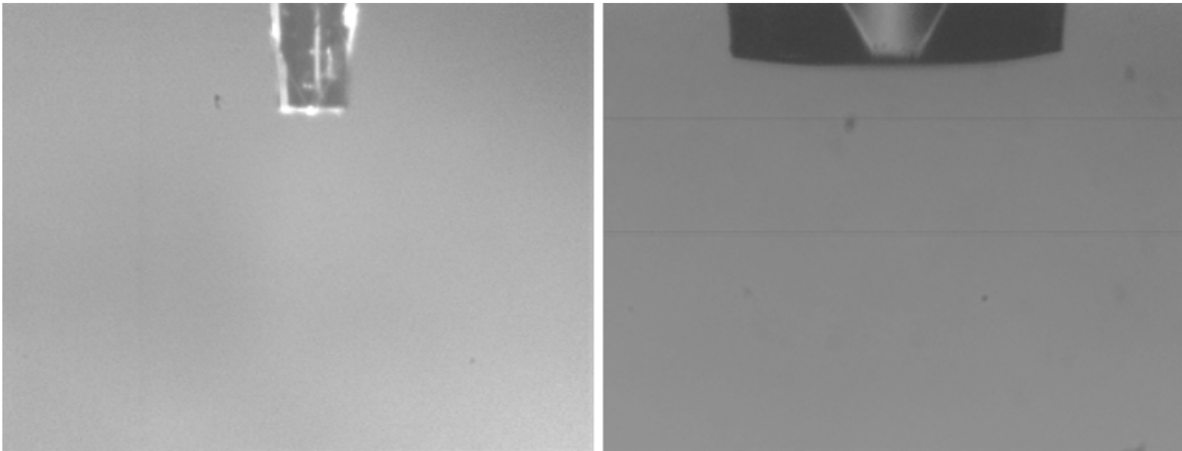
**Figure 4:** *The parameter list in the main window for a controller with a choice of pulse shapes, with “arbitrary waveform” selected, MicroFab Technologies, Inc., User’s Guide, June 2012, Version 4.1.*

I assume for the purpose of the ensuing step-by-step example that the JetServer™ software is already active. My objective is to outline the "mechanical" stages involved in fine-tuning a device's jetting behavior. As a result, the method for choosing specific parameters is not covered in great detail.

1. If multiple jets or dispensing channels are available, I make sure that only the one jet or channel to be worked on is showing a green device; click on its button if it is not. If then any other device icons still show green, click on each of those to make them change color to dark blue. Note that, depending on the hardware components in your setup, the number of device icons shown may vary.



2. I make sure the device is located over a waste bucket.
3. Move the jet or the horizontal camera until its tip appears in the field of view of the camera. Adjust the focus of the camera to obtain a sharp image of the tip, in particular the vertical sides of it.



**Figure 5:** *Jetting device tip in view of the horizontal camera for controlling jet formation during pulse shape tuning, MicroFab Technologies, Inc., User's Guide, June 2012, Version 4.1.*

Figure 5's two photos show the range of magnifications that can be used to adjust the pulse shape for jetting devices. The front faces of the devices in the image have a diameter of roughly  $550\mu\text{m}$ . Both times, it has been easier to see the device tip in the field of view using external light sources, such as a flashlight. If no readily accessible source exists, you can think about activating the main window's jetting device by pulsing it: Near the top center, click the Start Jet Continuous button; click it again to stop. This will activate the strobe light, illuminating the desired region in front of the camera (and may generate droplets already, too).

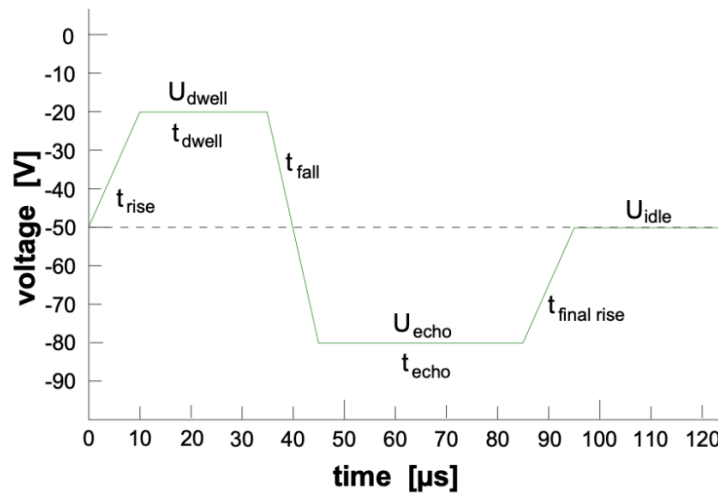
In rare instances, the camera may have a reticle projector that projects a cross with scale divisions in the middle to aid in estimating distances when calculating drop velocity. On more recent MicroFab dispensing test systems, the camera image is captured using a frame grabber and shown in a separate window on the computer screen; in this case, the display program can overlay

a vertical scale.

4. In the main window, the columns of input fields on the left side include the pulse shape parameters. If you have a recommended set of numbers, type them into the corresponding fields. If you don't have any, start with the numbers shown in Figure 7, except that +20 V and -20 V are probably better dwell and echo voltages to begin with.

There are five times for “Rise 1”, “Dwell”, “Fall”, “Echo”, and “Rise 2” (also called final rise), and three voltage levels “Idle”, “Dwell” and “Echo” as illustrated in Figure 6. The second half of the pulse is called the echo and needs not be used. It can be turned off by setting the echo voltage equal to the idle voltage (The total pulse length still includes the echo and final rise times).

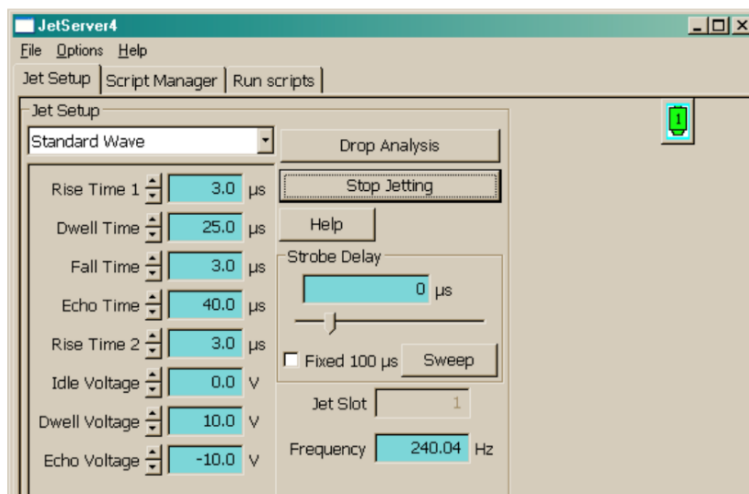
With MicroFab's jetting devices and drive electronics, the echo part is used more often than not, the most notable exceptions being the dispensing of isopropanol and sometimes water and water-based solutions.



**Figure 6:** Principle of pulse shapes used to fire jetting devices, MicroFab Technologies, Inc., User's Guide, June 2012, Version 4.1.

5. Click on the Start Jet Continuous button. It will change to display now Stop Jetting and the jetting device will be continuously pulsed at the frequency given further down below the Stop Jetting button (Figure 7).

6. If the parameters are suitable for the fluid being used, formation of droplets should occur. To find out, the trigger to the strobe light needs to be delayed relative to the pulse sent to the jetting device. This can be accomplished with the slider for Strobe delay in the main window, an adjustment knob on the jet controller, or a potentiometer driven delay in the MicroFab strobe drive unit, which is either a separate box or integrated in a pneumatics console. As a rule of thumb, the drop formation at the orifice begins some time around the end of the fall in the pulse shape.
7. If no drop formation is present, or if it is not satisfactory, change some pulse shape parameter and press the “Enter” key. The jetting will be stopped, the changed pulse shape sent to the controller, and the jetting resumed automatically.
8. Another way to influence the drop formation that is useful and often required but not necessarily controlled through the JetServer™ program is the “back pressure”, i.e., pressure applied to the fluid in the reservoir. This can be done with the help of a separate pneumatic console or an optional computer-operated pressure controller.



**Figure 7:** Main program window with pulsing at the output going on. The trigger mode is set to continuous automatically when using the “Start Jet Continuous” button. Alternatively, the mode could be set directly in the “Trigger mode” box and the “Trigger Jet” button underneath it used to start pulsing, MicroFab Technologies, Inc., User’s Guide, June 2012, Version 4.1.

The two main purposes of changing the back pressure are as follows: The device can first be cleaned by either using a lot of pressure to flood liquid through it or a lot of vacuum to draw material out of the orifice and into the reservoir or the filter between the reservoir and the device.

To prevent the fluid from dripping or running out of the orifice spontaneously, a small positive back pressure (measured in psi, from fractions to a very few) or negative pressure (measured in cm of water, range of a few; in psi, a fraction) can be applied gradually.

9. Repeat the trials described in steps 4-8 until good drop formation is observed.

In many cases, a useful strategy for searching for good pulse shape parameters is to concentrate on the dwell voltage and dwell time. With an echo used, it is common, although not necessary, to keep the echo voltage the negative of the dwell voltage relative to the idle voltage.

Change one parameter at a time in steps of very roughly 10% (e.g., 2 V or 2  $\mu$ s for standard starting numbers suggested above in step 4). If satellites appear, a reduction of the dwell voltage as well as changes in the dwell and echo times can help to reduce or prevent their occurrence.

10. To stop the pulsing, click on the Stop Jetting button. It will revert to Start Jet Continuous .

11. To permanently save a good set of parameters, open the File menu and click on Save Jetting Parameters, which will save the parameters to the last file that has been opened or saved (and always to JETLABDEVICE.DAT), or click on Save As Jetting Parameters to allow yourself to pick an existing or new name for the file.

12. If you use the Save As Jetting Parameters entry of the file menu, a standard Windows file save dialog will appear. Provide a file name and click on the Save button. The parameter set can be reloaded later by using the Open Jetting Parameters entry of the File menu. Note that these actions affect the parameters of all jets known at that time in the JetServer<sup>TM</sup> program.

13. After saving the parameters to a file, the program will automatically return to the main window.

### **2.2.3 Standard Microdispensing Procedure**

1. Unpack the syringe.
2. Dip the syringe into the liquid and suck it up.
3. Put on liquid filter at the top of syringe.
4. Put syringe with filter onto liquid reservoir and press liquid slowly through filter into the liquid reservoir.
5. Load filtered liquid into dispenser head using a pipette.
6. Wipe nozzle tip with lint-free tissue if excess liquid runs out of it.
7. Push down dispenser head gently into the holder using two fingers.
8. Ensure the power that the laser receives from the power supply is set at 0.1W.
9. Set standard settings on JetServer program.
10. Set 'Trigger Source' on JetServer program to Internal.
11. Set 'Trigger Mode' on JetServer program to Single.
12. Hit 'Trigger Jet' on JetServer program.
13. Liquid runs out of the nozzle of the dispenser head as a smooth jet.
14. To check whether a droplet is visible, 'Strobe delay' can be activated. A stable droplet is visible by Strobe delay = 500 $\mu$ s.

Note:

- Be careful that no particles will contaminate the dispenser head.
- Flush out dispenser head with distilled water after experiment is completed.

## **2.3 Using an Electrodynamic Balance (EDB) to Study High Total Dissolved Salt Microdroplets**

In an electrodynamic balance, a charged particle with a usual radius of 2 to 10  $\mu\text{m}$  is levitated.<sup>7</sup> Using automatic mass flow controllers, the lab air/H<sub>2</sub>O ratio of steady gas flow is adjusted to modulate the RH. In an experiment, the temperature is dictated by lab conditions while the relative humidity is altered gradually. The sensor is immediately calibrated in the trap using the relative humidity of various salts during deliquescence. Between 10%RH and 80%RH and above 80%RH, its precision is  $\pm 1.5\%$ RH and  $\pm 3\%$ RH, respectively.

A Nd:YAG laser (532 nm) illuminates the particle from below in order to visualize it. The DC voltage is adjusted to account for the gravitational force using an automated feedback loop and a video image of the particle captured on a CCD detector.<sup>33</sup> Therefore, a shift in DC voltage can be used as a direct indicator of a mass change. We can determine the mass growth factor if the voltage under dry conditions (RH<10%) correlates to the particle's dry mass ( $M_{\text{dry}}$ ).

### **2.3.1 EDB Operation**

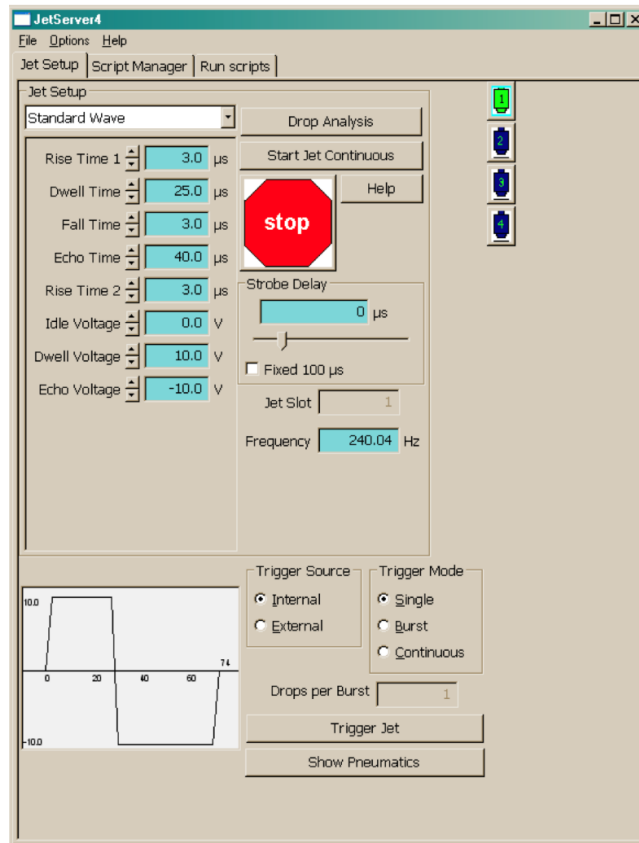
An electrodynamic balance can be used to suspend a micron-sized charged particle in air within a narrow region. A Millikan type DC field acting against gravitation is layered here with a non-uniform alternating field with a concentrating effect. In the null point of the non-uniform field, the charged particle is retained in space. The direct current field is applied between two endcap electrodes, and the alternating field is applied to two electrodes above and below the midplane.

The balance is housed in a three-walled glass chamber with a cooling liquid flowing through the inner walls and a vacuum isolation between the outer walls. The chamber is continually pumped with a constant flow of an appropriate gas mixture, and the total pressure can be changed between 150 and 1000 hPa.<sup>33</sup>

## 2.4 Software Used

This section contains experimental data pertaining specific as well as adjusted settings used to run two software programs namely, the JetServer program and EDB software (LabView) with the aim of achieving a goal which is to trap levitated droplets in both chambers (Top Balance and Bottom Balance) of the EDB device. The dispenser should not make droplets that are too small in order to show up on the camera.

Below is a distinct image showing the interface of one of the software programs that I used whilst performing my experiments in the lab.



**Figure 8:** *JetServer4 Program Interface, MicroFab Technologies, Inc., User's Guide, June 2012, Version 4.1.*

All accessory devices needed for the EDB device to generate droplets must be rightly calibrated, operated and functioning in order for these two software programs to run effectively. For all experiments carried out so far, the main solution used has been sodium chloride (NaCl),

and a saved (default) setting for the JetServer4 application was used for trapping a  $\sim 20\mu\text{m}$  droplet of 5wt% NaCl.

#### 2.4.1 JetServer4 & EDB Software Settings

STANDARD (DEFAULT) JETSERVER4 SETTINGS	
Jet Setting Name	Saved Settings
Rise Time 1	3 $\mu\text{s}$
Dwell Time	25 $\mu\text{s}$
Fall Time	3 $\mu\text{s}$
Echo Time	40 $\mu\text{s}$
Rise Time 2	3 $\mu\text{s}$
Idle Voltage	0V
Dwell Voltage	24V
Echo Voltage	-20V
Strobe Delay	0 $\mu\text{s}$
Frequency	240Hz

**Table 1:** Standard JetServer setting used for 5wt% NaCl.

#### 2.4.2 Target

Merging two droplets for a 5wt% NaCl with 50 $\mu\text{m}$  capillary. A single droplet got trapped with no accompanying satellite in both the Top and Bottom Balance which was finally merged successfully as a single droplet with specific settings used.

Note:

- I utilized MicroLev LLC's proprietary LabView software for experimental data collection and control (especially with the voltage).



<b>TOP BALANCE SETTING</b>			
<b>JetServer4</b>		<b>EDB</b>	
Rise Time 1	3 $\mu$ s	Induction Voltage	-350V
Dwell Time	25 $\mu$ s	Top Balance	125V
Fall Time	9 $\mu$ s	Bottom Balance	0V
Echo Time	40 $\mu$ s	Amplitude	500m
Rise Time 2	3 $\mu$ s	Frequency	300Hz
Idle Voltage	0V	Total Flow	60sccm
Dwell Voltage	14V	Wet Flow	60%
Echo Voltage	-20V	Dry Flow	40%
Frequency	240Hz		

**Table 2:** Top Balance setting used for 5wt% NaCl.

<b>BOTTOM BALANCE SETTING</b>			
<b>JetServer4</b>		<b>EDB</b>	
Rise Time 1	3 $\mu$ s	Induction Voltage	150V
Dwell Time	25 $\mu$ s	Top Balance	0V
Fall Time	3 $\mu$ s	Bottom Balance	-150V
Echo Time	40 $\mu$ s	Amplitude	500m
Rise Time 2	3 $\mu$ s	Frequency	300Hz
Idle Voltage	0V	Total Flow	60sccm
Dwell Voltage	18V	Wet Flow	60%
Echo Voltage	-20V	Dry Flow	40%
Frequency	240Hz		

**Table 3:** Bottom Balance setting used for 5wt% NaCl.

Certain observations were made, and comments noted whiles conducting these experiments to get a single droplet merged from two droplets. Amongst them are:

- Feedback button on EDB software program (LabView) automatically adjusts Bottom balance once a droplet is loaded there.
- Reduction of dwell voltage (by 2V) as well as changes of dwell and echo time can help reduce satellite generation.

- Change in frequency value has no impact on droplet size.
- A single trigger mode was used to get trapped droplets.
- If the droplet's image is offset, adjust the Bottom voltage for a centered image.

In the next chapter, I discuss a Theoretical Background as a method for relating the measured voltage to the droplet mass in a sensitive fashion.

### 3.0 THEORETICAL BACKGROUND

#### 3.1 Force balance: gravity, charge, and drag

The main reason for deriving the force balance is to comprehend the dynamical behavior of levitated multi-drop and single-drop systems. The forces that are acting on the levitated drops must be included in the simulation of the system in order to do this. The principal forces include the gravitational force, the force of mutual Coulombic repulsion, the force of dissipation caused by viscous drag, and the dielectrophoretic force resulting from the interaction of the droplet's internal polarization with the spatial gradient of the electric field.

Here, force balance as used in the water activity paper by Liang et al.<sup>26</sup> will be described mathematically, and details of how it can apply to data gathered during NaCl experiment will be stated.

If there is no relative motion between the particle and the ambient gas, the electrostatic force will be equal to the weight of the particle. The mass of the particle,  $m$ , can then be determined by:

$$mg = \frac{c_o q}{z_o} V_{bal} \quad (3.1)$$

where  $c_o$ , is a balance calibration constant,  $q$  is the number of electrostatic charges carried by the particle,  $V_{bal}$  is the dc voltage required to balance the particle,  $g$  is the gravitational constant, and  $z_o$ , is the distance between the top and bottom electrodes.

A new equation emerges if a drag force ( $F_D$ ) is included in the force balance on a particle with air flow:

$$mg = \frac{c_o q}{z_o} V_{bal,drag} - 6\pi\mu ua \quad (3.2)$$

where  $a$  is the radius of a known particle size for calibration, typically polystyrene latex (PSL) particles, and  $u$  is the approach velocity of the particle which is taken to be the velocity at the null

point of the EDB. At 20°C, the viscosity of air stream,  $\mu$ , is 1.82 X Pa.s<sup>27</sup>.

$$F_D = 6\pi\mu ua \text{ [In the EDB, at rest]} \quad (3.3)$$

The equation can be re-written as;

$$mg = \frac{c_o q}{z_o} V_{bal,drag} - F_D \quad (3.4)$$

Also,

$$u = (V_p - V_{air}) \quad (3.5)$$

where  $V_p$  is the velocity of the particles and  $V_{air}$  is the velocity of air, so  $u$  is the relative velocity between particles and air.

$$\text{In the balance, } V_p = 0 \quad (3.6)$$

Hence,

$$F_D = 6\pi\mu V_{air} a \quad (3.7)$$

Furthermore,

$$\dot{m}_{air} = \rho_{air} A V_{air} = \rho_{air} Q \quad (3.8)$$

where  $\dot{m}_{air}$  is the mass flow rate of air,  $\rho_{air}$  is the density of air,  $A$  is the cross-sectional area of the dispenser tube and  $Q$  is the volumetric flow rate.

$$V_{air} = \frac{\dot{m}_{air}}{\rho_{air} A} = Q \quad (3.9)$$

$$Q = A V_{air} ; V_{air} = \frac{Q}{A} \quad (3.10)$$

Mass of particle,

$$m = \rho_p V_p \quad (3.11)$$

$$m = \rho_p \left( \frac{4}{3} \pi r_p^3 \right) = \rho_p \left( \frac{4}{3} \pi a^3 \right) \quad (3.12)$$

where  $\rho_p$  is the density of the particles and  $r_p$  is the radius of the particles.

$$3m = \rho_p 4\pi a^3 \quad (3.13)$$

$$a^3 = \frac{3m}{\rho_p 4\pi} \quad (3.14)$$

$$a = \left( \frac{3m}{\rho_p 4\pi} \right)^{\frac{1}{3}} \quad (3.15)$$

Putting  $a = \left( \frac{3m}{\rho_p 4\pi} \right)^{\frac{1}{3}}$  into  $mg = \frac{c_o q}{z_o} V_{bal,drag} - 6\pi\mu u a$  gives;

$$mg = \frac{c_o q}{z_o} V_{bal,drag} - 6\pi\mu u \left( \frac{3m}{\rho_p 4\pi} \right)^{\frac{1}{3}} \quad (3.16)$$

$$m_o g = m_o g \frac{V_{bal,drag,0}}{V_{bal,0}} - 6\pi\mu u \left( \frac{3m_o}{\rho_p 4\pi} \right)^{\frac{1}{3}} \quad (3.17)$$

$$mfs_o = \frac{m_{dry}}{m_o} \quad (3.18)$$

where  $V_{bal,drag,0}$  and  $V_{bal,0}$  are the balancing dc voltages of the particles equilibrated at RH<sub>1</sub>, with and without the air flow are measured,  $m_o$  is the initial mass of the droplet,  $mfs_o$  is the initial mass fraction of solute and  $m_{dry}$  is the dry mass of the particle.

The density,  $\rho$ , is a function of mfs and can be obtained from literature data<sup>27</sup> or approximated from an established mixing rule from the literature.<sup>28</sup>

### 3.1.1 Droplet mass measurements

$$mg = \frac{c_o q}{z_o} V_{bal,drag} - 6\pi\mu u \left( \frac{3m}{4\pi\rho_p} \right)^{\frac{1}{3}} \quad (3.1.1)$$

Multiplying through the equation by the denominator  $[(4\pi\rho_p)^{\frac{1}{3}}z_o]$  gives;

$$(4\pi\rho_p)^{\frac{1}{3}}z_0mg = (4\pi\rho_p)^{\frac{1}{3}}c_0qV_{bal,drag} - 6\pi\mu(3m)^{\frac{1}{3}}z_0 \quad (3.1.2)$$

Grouping like terms gives;

$$(4\pi\rho_p)^{\frac{1}{3}}c_0qV_{bal,drag} = (4\pi\rho_p)^{\frac{1}{3}}z_0mg + 6\pi\mu(3m)^{\frac{1}{3}}z_0 \quad (3.1.3)$$

Dividing both sides of the equation by  $[(4\pi\rho_p)^{\frac{1}{3}}c_0q]$  reduces the equation to;

$$V_{bal,drag} = \frac{z_0mg}{c_0q} + \frac{6\pi\mu(3m)^{\frac{1}{3}}z_0}{(4\pi\rho_p)^{\frac{1}{3}}c_0q} \quad (3.1.4)$$

Re-arranging the terms in the equation gives;

$$V_{bal,drag} = \frac{z_0mg}{c_0q} + \frac{6\pi\mu z_0}{c_0q} \left( \frac{3m}{4\pi\rho_p} \right)^{\frac{1}{3}} \quad (3.1.5)$$

This derivation will be used to interpret the data described in the Results and Discussion section of this document, and all future data. It will also be useful in determining the water activity and hygroscopicity of levitated microdroplets in the electrodynamic device.

## 4.0 RESULTS AND DISCUSSION

### 4.1 Initial tests of hygroscopic behavior of NaCl particle

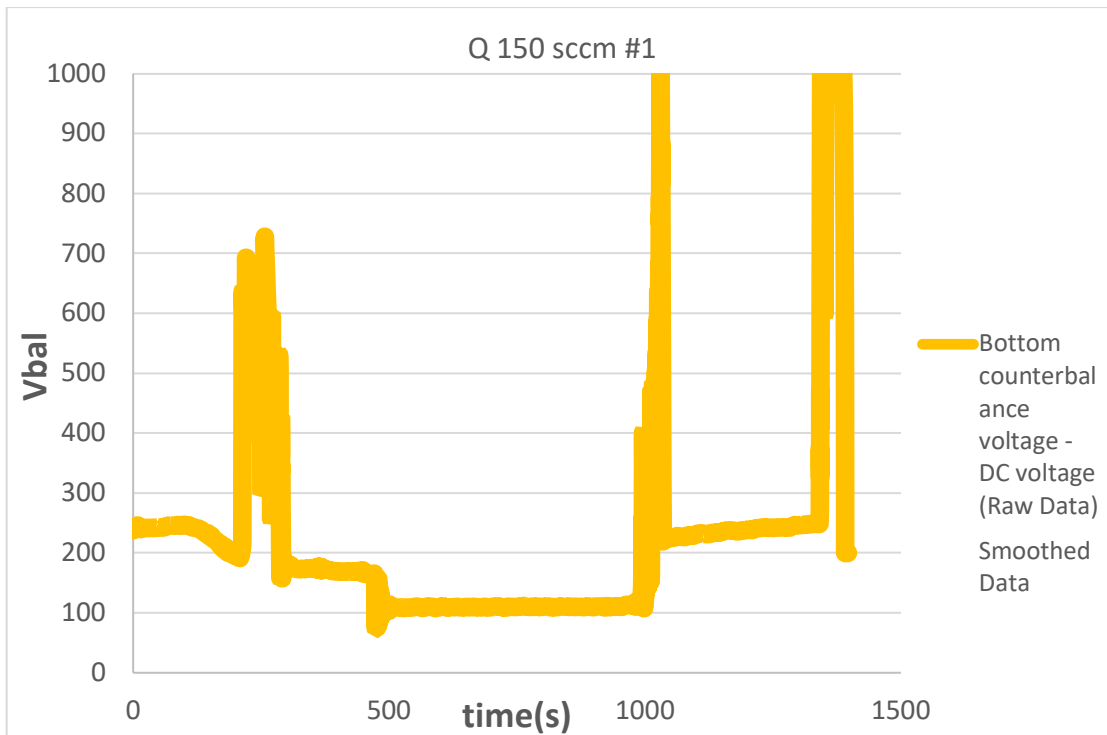
Five different droplets were studied, all containing 5wt% NaCl. Table 4 contains the experimental conditions. The two parameters that I varied were the volume flow rate the droplet is subjected to and the wet/dry ratio. Varying the wet/dry ratio serves the purpose of changing the droplet mass due to loss of water vapor (evaporation) or uptake of water vapor (condensation). Appendix A contains the tables with the raw and smoothed experimental data.

The particle size stayed constant throughout the humidifying process up to  $RH = 65.5\%$ , at which point a slight reduction in size was noticed due to water adsorption in the solid salts' lattice flaws and structural rearrangement inside the crystal lattice. Note that I am estimating RH from the wet/dry flow ratio. From  $RH = 67.7$  to  $68.0\%$ , a first deliquescence transition was observed, and its size significantly grew. It grew steadily as RH was raised until  $RH = 73.7\%$ , where the second transition took place. The particle then kept growing as the RH was raised further.

As the RH is raised from 0 to  $67.7\%$ , the morphology does not significantly change. However, the particle boundaries start to round at  $73.7\%$  RH. It deliquesces at  $76\%$  RH and transforms into a solution droplet with a distinct deliquescence sphere. The deliquescence sphere contracts when the RH is decreased from  $76\%$  till the ERH of  $44\%$  is obtained. It then recrystallizes at this point. This experiment enables a comparison between a pure NaCl particle's hygroscopic behavior and cycle in the EDB device at a specific flow rate, first with noise signals and then with minimum noise signals as seen in Figure 11.

Droplet #	Q (sccm)	Vbal/eff start	Vbal/eff end	Vbal/del start	Vbal/del end	Uptake time (min)	Induction voltage	Vel. of air (m/s)	Calculated Q (sccm)
1	150	158	105	110	248	8.05	-150	0.0789	150
	200	264	147	147	282	4.633333333	-150	0.1053	200
	250	278	202	207	387	3.266666667	-150	0.1316	250
2	150	161	101	111	214	18.18333333	-150	0.0789	150
	200	207	145	140	278	12.55	-150	0.1053	200
	250	265	197	208	380	6.2	-150	0.1316	250
	275	314	219	252	445	5.716666667	-150	0.1447	275
	300	391	284	291	1000	1.933333333	-150	0.1579	300
3	150	168	119	126	497	5.666666667	-150	0.0789	150
4	200	0	0	126	40	2.1	-250	0.1053	200
5	200	0	0	268	1000	3.8	-100	0.1053	200

**Table 4:** Data used to analyze the hygroscopicity cycle of different levitated NaCl droplets in the EDB device at specific flow rates during the conduction of the NaCl deliquescence/efflorescence experiment.



**Figure 9:** Comparing the hygroscopicity cycles of NaCl droplet #1 in the EDB device at a flow rate of 150sccm (1) With noise signals (2) With minimum noise signals.



## 5.0 CONCLUSION AND FUTURE WORK

Efflorescence and deliquescence (defined in Appendix C) and their hysteresis occur in NaCl salt (an inorganic compound) when levitated inside the EDB at specific settings. Since RH was only estimated from the ratio of wet/dry flow rates, these experiments should be repeated with an RH probe in the chamber. The study of the phase and morphology of various hydrocarbon petrochemicals in inorganic and organic microdroplets using the EDB is possible due to demonstrating that this unique DBET setup and the force balance development in Chapter 3 enables collection of new data on these critical and multi-component solutions. Hence more experimental data is required for further research that will benefit the energy sector.

To be able to draw more definitive conclusions about the results obtained regarding hygroscopicity when these microdroplets are levitated inside the EDB at specific settings, research and additional experiments on other inorganic compounds ( $\text{CaCl}_2$ ,  $\text{MgCl}_2$ ,  $\text{NaBr}$ ,  $(\text{NH}_4)_2\text{SO}_4$ ,...), as well as organic compounds (Citric acid, Glutaric acid,...) should be carried out. Elimination of coagulated hydrocarbons/petrochemicals in organic and inorganic aqueous solutions with the long-term goal of guiding new technologies is required for treating and recycling produced water effluent sustainably.

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## APPENDIX A

### Data Analysis: Hygroscopicity cycles of 5wt% NaCl droplet

Time - DC voltage	Bottom counterbalance voltage - DC voltage (Raw Data)	Smoothed Data	Before Efflorescence (Smoothed Data)
		DF	DF
0	237.9	237.9	237.9
0.05	237.9	237.9	237.9
0.1	237.9	237.9	237.9
0.15	237.9	237.9	237.9
0.199	237.8	237.899985	237.89997
0.249	237.8	237.89997	237.89994
0.299	237.8	237.899955	237.89991
0.35	237.8	237.89994	237.8998801
0.4	237.7	237.89991	237.8998201
0.45	237.7	237.89988	237.8997601
0.5	237.7	237.8998501	237.8997002
0.55	237.7	237.8998201	237.8996403
0.599	237.6	237.8997751	237.8995504
0.649	237.6	237.8997301	237.8994605
0.699	237.6	237.8996852	237.8993707
0.75	237.6	237.8996402	237.8992809
0.8	237.5	237.8995803	237.8991611
0.85	237.5	237.8995203	237.8990414
0.9	237.5	237.8994604	237.8989217
0.949	237.5	237.8994005	237.898802
0.999	237.5	237.8993406	237.8986823
1.049	237.5	237.8992807	237.8985627
1.1	237.5	237.8992208	237.8984432
1.15	237.5	237.8991609	237.8983236
1.2	237.5	237.899101	237.8982041
1.25	237.5	237.8990412	237.8980847
1.3	237.5	237.8989813	237.8979652
1.349	237.6	237.8989365	237.8978759
1.399	237.6	237.8988916	237.8977865
1.449	237.7	237.8988618	237.8977272
1.5	237.7	237.898832	237.8976678
1.55	237.7	237.8988021	237.8976085
1.6	237.8	237.8987873	237.8975793
1.65	237.8	237.8987725	237.89755
1.699	237.8	237.8987577	237.8975207
1.749	237.9	237.8987579	237.8975215
1.799	237.9	237.8987581	237.8975222
1.85	237.9	237.8987582	237.8975229
1.9	237.9	237.8987584	237.8975237
1.95	237.9	237.8987586	237.8975244
2	237.9	237.8987588	237.8975252
2.05	237.9	237.898759	237.8975259

2.099	237.9	237.8987592	237.8975267
2.149	237.9	237.8987594	237.8975274
2.199	237.9	237.8987595	237.8975281
2.25	237.9	237.8987597	237.8975289
2.3	237.9	237.8987599	237.8975296
2.35	237.9	237.8987601	237.8975304
....	....	....	....
476.6	141.3	238.063596	221.409255
476.65	145.3	238.0496815	221.3870925
476.7	146.1	238.035889	221.3645064
476.749	147	238.0222336	221.342197
476.799	147.8	238.0087003	221.3201344
476.849	148.6	237.995289	221.2983183
476.899	145.2	237.9813697	221.2754888
476.949	141.8	237.9669425	221.2516462
477	138.5	237.9520225	221.2268207
477.049	133.4	237.9363397	221.2004726
477.099	128.3	237.9198942	221.1726025
477.149	123.2	237.9026862	221.1432107
477.199	117.8	237.8846708	221.1122078
477.249	112.5	237.8658631	221.0796241
477.299	107.2	237.8462632	221.0454602
477.35	102.1	237.8259013	221.0097766
477.4	97	237.8047774	220.9725736
477.449	91.9	237.7828917	220.9338519
477.499	88.5	237.7604993	220.8941217
477.549	85.1	237.7376002	220.8533835
477.599	81.6	237.7141796	220.8116075
477.649	78.2	237.6902524	220.768824
477.699	77.6	237.6662389	220.7258733
477.75	77	237.642139	220.6827556
477.799	76.4	237.6179526	220.6394707
477.849	79	237.5941599	220.5969789
477.899	81.5	237.5707458	220.5552498
477.949	84	237.5477102	220.5142832
477.999	89.8	237.525548	220.4750689
478.049	95.5	237.5042442	220.4375764
478.1	101.3	237.4838136	220.4018351
478.15	108.6	237.464481	220.3682946
478.199	115.8	237.4462313	220.3369241
478.249	123	237.4290644	220.307723
478.299	129.6	237.41289	220.2805107
478.349	136.2	237.3977081	220.2552866
478.399	142.9	237.3835335	220.23208
478.449	149.5	237.3703509	220.2108603
478.5	152.4	237.3576054	220.1905171
478.549	155.3	237.3452967	220.1710499

**Table 5:** Data used to analyze the hygroscopicity cycle NaCl droplet #1 in the EDB device at 150sccm flow rate during the conduction of the NaCl deliquescence/efflorescence experiment (1) With noise signals (2) With minimum noise signals, before efflorescence occurs.



<b>Time - DC voltage</b>	<b>Bottom counterbalance voltage - DC voltage (Raw Data)</b>	<b>Smoothed Data</b>	<b>Before Efflorescence (Smoothed Data)</b>	<b>During Efflorescence (Smoothed Data)</b>
				<b>DF</b>
478.599	158.2	237.3334249		158.2
478.649	156.5	237.3212999		158.183
478.699	154.7	237.3089067		158.14817
478.749	153	237.2962604		158.0966883
478.799	148.2	237.2828959		157.9977214
478.85	143.4	237.2688135		157.8517442
478.9	138.6	237.2540132		157.6592268
478.949	132.6	237.2383151		157.4086345
478.999	126.6	237.2217193		157.1005481
479.049	120.6	237.2042261		156.7355427
479.099	114.7	237.1858505		156.3151872
479.149	108.8	237.1665926		155.8400354
479.199	102.9	237.1464526		155.310635
479.25	97	237.1254306		154.7275287
479.3	92	237.1036618		154.1002534
479.349	87	237.0811463		153.4292508
479.399	82	237.0578841		152.7149583
479.449	83.3	237.0348204		152.0208088
479.499	84.5	237.0119402		151.3456007
479.549	85.8	236.9892584		150.6901447
479.6	85.4	236.96652		150.0372432
479.65	85.1	236.94374		149.3878708
479.699	84.7	236.9209035		148.7409921
479.749	89.4	236.8987753		148.1475822
479.799	94.1	236.8773555		147.6071063
479.849	98.8	236.8566439		147.1190353
479.899	104.5	236.8367904		146.6928449
479.949	110.2	236.8177949		146.3279165
480	115.9	236.7996572		146.0236373
480.049	121.6	236.7823773		145.7794009
480.099	126.5	236.7658349		145.5866069
480.149	131.5	236.750045		145.4457408
480.199	136.4	236.7349925		145.3552834
480.249	139.7	236.7204373		145.2987306
480.299	143	236.7063792		145.2757433
480.35	146.2	236.6928033		145.2849859
480.4	145.6	236.6791393		145.288136
480.449	144.9	236.6653725		145.2842546
480.499	144.2	236.6515027		145.2734121
480.549	140.8	236.6371249		145.228678
480.599	137.3	236.6222244		145.1493912
480.649	133.9	236.606816		145.0368973
480.699	128.9	236.59066		144.8755283
480.75	123.9	236.5737564		144.665773
480.799	119	236.5561203		144.4091153

480.849	114	236.5377369		144.1050241
480.899	108.7	236.5185613		143.7509739
480.949	103.4	236.4985935		143.3474642
480.999	98.1	236.4778337		142.8949895
481.049	93.8	236.456432		142.4040396
481.1	89.6	236.4344036		141.8759992
481.149	85.3	236.4117334		141.3102392
481.199	86.6	236.3892616		140.7631368
481.249	88	236.3670032		140.2355055
481.299	89.3	236.3449432		139.7261504
481.349	93.6	236.3235315		139.2648889
481.399	97.8	236.3027529		138.85024
481.449	102.1	236.2826225		138.4827376
481.5	105.2	236.2629601		138.1499103
481.549	108.4	236.2437807		137.8524112
481.599	111.5	236.2250691		137.588887
481.649	114.7	236.2068403		137.3599982
481.699	117.6	236.1890493		137.1623982
481.749	120.4	236.171681		136.9947742
481.799	123.3	236.1547502		136.8578265
481.85	126.3	236.138272		136.7522482
481.899	129.2	236.1222313		136.6767257
481.949	132.2	236.1066429		136.6319585
481.999	131.8	236.0909969		136.5836389
482.049	131.5	236.0753083		136.5328025
482.099	131.2	236.059577		136.4794745
482.149	129.4	236.043578		136.4086797
482.199	127.7	236.0273265		136.3215929
482.25	126	236.0108224		136.218377
482.299	122.9	235.9938558		136.0851932
482.349	119.8	235.9764267		135.9223413
482.399	116.8	235.9585502		135.7311179
482.449	113.7	235.9402115		135.5108067
482.499	109.9	235.9213054		135.2546986
482.549	106.1	235.9018322		134.9631516
482.6	102.4	235.881807		134.6375201
482.649	99.5	235.8613497		134.2861449
482.699	96.6	235.8404605		133.9092835
482.749	93.8	235.8191544		133.5081906
482.799	92.1	235.7975965		133.0941087
482.849	90.5	235.7758019		132.6681677
482.899	88.9	235.7537705		132.230486
482.95	89.2	235.7317875		131.8001811
483	89.5	235.7098527		131.3771793
483.049	89.8	235.6879662		130.9614075
483.099	94.4	235.666773		130.5957934
483.149	99	235.646273		130.2798355
483.199	103.7	235.6264811		130.0140371
483.249	108.3	235.6073821		129.7968968
483.299	112.2	235.588871		129.6209278
483.35	116.2	235.5709627		129.4867185

483.399	120.1	235.553642		129.3928513
483.449	124	235.536909		129.3389228
483.499	127.8	235.5207484		129.3235336
483.549	131.6	235.5051603		129.3462983
483.599	130	235.4893345		129.3528353
483.649	128.3	235.4732561		129.3423069
483.699	126.7	235.4569402		129.3158839
483.75	126.5	235.4405966		129.287725
483.799	126.2	235.4242105		129.2568478
483.849	125.9	235.4077819		129.2232793
483.899	121.8	235.3907407		129.1490465
483.949	117.7	235.3730871		129.034556
483.999	113.6	235.3548212		128.8802105
484.049	109.5	235.3359429		128.6864084
484.1	106.4	235.3166025		128.4635443
484.15	103.2	235.296785		128.2109088
484.199	100.1	235.2765055		127.9297998
484.249	98.9	235.2560491		127.6395018
484.299	97.6	235.2354006		127.3391067
484.349	96.4	235.2145753		127.0297157
484.399	94.7	235.1934982		126.7064185
484.45	93	235.1721691		126.3693543
484.5	91.3	235.1505883		126.0186608
484.549	93.2	235.1292957		125.6904742
484.599	95.2	235.1083063		125.3855694
484.649	97.1	235.0876051		125.1027137
484.699	99.3	235.0672369		124.8446866
484.749	101.4	235.0471868		124.6102397
484.799	103.6	235.0274698		124.4001373
484.85	105.7	235.0080706		124.213136
484.899	109.4	234.9892294		124.0650046
484.949	113	234.9709311		123.9543546
484.999	116.7	234.9531904		123.881811
485.049	118.2	234.9356774		123.8249929
485.099	119.8	234.9184071		123.784743
485.149	121.3	234.9013643		123.7598955
485.199	121	234.8842791		123.7322966
485.25	120.7	234.8671515		123.7019736
485.299	120.3	234.8499664		123.6679539
485.349	120.6	234.8328289		123.6372744
485.399	120.8	234.815724		123.6089016
485.449	121.1	234.7986666		123.5838126
485.499	118.5	234.7812218		123.5329745
485.549	115.9	234.7633896		123.4566447
485.6	113.3	234.7451701		123.3550783
485.649	110.7	234.7265634		123.2285275
485.699	108.8	234.7076744		123.0842422
485.749	106.9	234.6885032		122.9223998
485.799	105.1	234.6690649		

**Table 6:** Data used to analyze the hygroscopicity cycle of NaCl droplet #1 in the EDB device at 150sccm flow rate during the conduction of the NaCl deliquescence/efflorescence experiment (1) With noise signals (2) With minimum noise signals, during which efflorescence occurs.

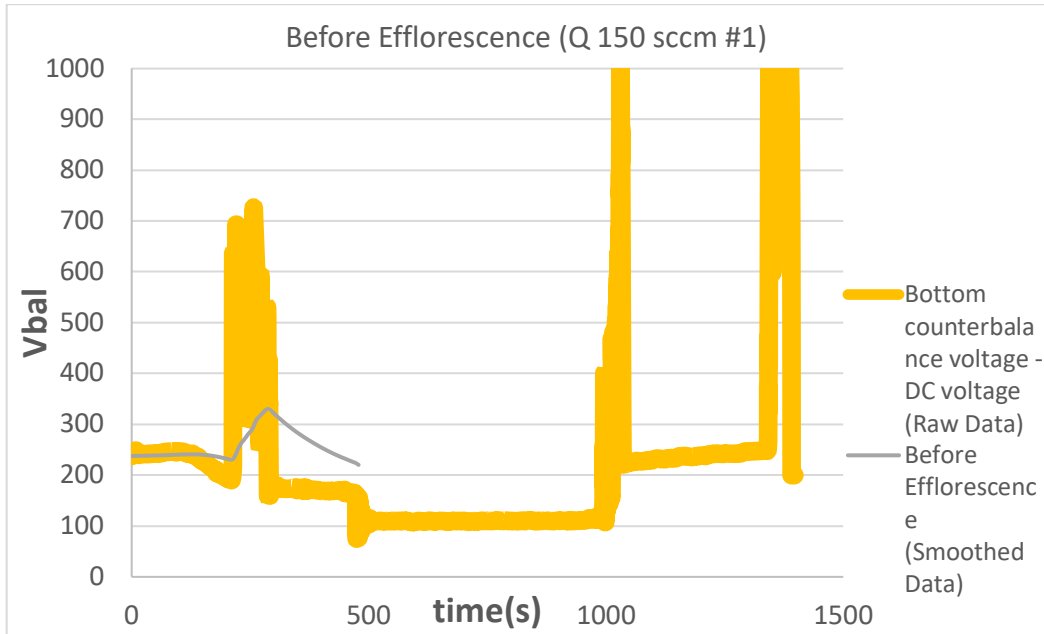
<b>Time - DC voltage</b>	<b>Bottom counterbalance voltage - DC voltage (Raw Data)</b>	<b>Smoothed Data</b>	<b>Before Efflorescence (Smoothed Data)</b>	<b>During Efflorescence (Smoothed Data)</b>	<b>After Efflorescence (Smoothed Data)</b>
					<b>DF</b>
485.849	103.9	234.6494496			103.9
485.899	102.8	234.6296722			103.89967
485.949	101.7	234.6097327			103.8990101
486	100.4	234.5896013			103.8979604
486.049	99.1	234.5692778			103.896521
486.099	97.7	234.5487474			103.8946621
486.149	99.5	234.5284901			103.8933437
486.199	101.4	234.5085208			103.8925956
486.249	103.2	234.4888246			103.8923879
486.304	105	234.4694012			103.8927202
486.35	106.8	234.4502508			103.8935923
486.399	108.6	234.4313733			103.8950043
486.449	110.5	234.4127836			103.8969858
486.499	112.5	234.3944967			103.8995667
486.549	114.4	234.3764975			103.9027168
486.599	116.4	234.358801			103.906466
486.649	116.8	234.3411672			103.910334
486.699	117.1	234.323581			103.9142909
486.75	117.4	234.3060425			103.9183367
486.799	116.6	234.2883866			103.9221412
486.849	115.8	234.2706133			103.9257045
486.899	114.9	234.2527077			103.9289968
486.949	113.5	234.2345948			103.9318681
486.999	112	234.2162596			103.9342885
487.049	110.6	234.1977172			103.9362883
487.1	109.4	234.1789975			103.9379274
487.149	108.3	234.1601157			103.939236
487.199	107.1	234.1410567			103.9401842
487.249	106	234.1218355			103.9408022
487.299	105.5	234.1025422			103.9412699
487.349	105.1	234.0831919			103.9416175
487.399	104.7	234.0637844			103.9418451
487.45	105	234.0444248			103.9421625
487.499	105.2	234.0250981			103.9425399
487.549	105.5	234.0058194			103.9430071
487.599	106.5	233.9866935			103.9437742
487.649	107.4	233.9677055			103.9448111
487.699	108.4	233.9488703			103.9461476
487.749	109.3	233.930173			103.9477538
487.799	110.2	233.9116135			103.9496294
487.85	111.2	233.8932068			103.9518046
487.9	112	233.8749228			103.954219
487.949	112.9	233.8567765			103.9569027
487.999	113.8	233.838768			103.9598557
488.049	114.7	233.8208972			103.9630777

488.099	114.3	233.8029691			103.9661788
488.149	113.9	233.7849836			103.9691589
488.2	113.5	233.7669409			103.9720182
488.25	112.7	233.7487808			103.9746366
488.299	112	233.7305185			103.9770442
....	....	....			....
1395.049	200	312.0805042			412.9265489
1395.1	200	312.0636921			412.862671
1395.15	200	312.0468826			412.7988122
1395.2	200	312.0300756			412.7349725
1395.249	200	312.013271			412.671152
1395.299	200	311.9964691			412.6073507
1395.349	200	311.9796696			412.5435685
1395.399	200	311.9628726			412.4798054
1395.45	200	311.9460782			412.4160615
1395.5	200	311.9292863			412.3523367
1395.55	200	311.9124969			412.288631
1395.599	200	311.89571			412.2249444
1395.649	200	311.8789257			412.1612769
1395.699	200	311.8621438			412.0976285
1395.749	200	311.8453645			412.0339992
1395.799	200	311.8285877			411.970389
1395.85	200	311.8118134			411.9067979
1395.9	200	311.7950416			411.8432259
1395.95	200	311.7782724			411.7796729
1395.999	200	311.7615056			411.716139
1396.049	200	311.7447414			411.6526241
1396.099	200	311.7279797			411.5891284
1396.149	200	311.7112205			411.5256516
1396.2	200	311.6944638			411.4621939
1396.25	200	311.6777097			411.3987553
1396.3	200	311.660958			411.3353356
1396.349	200	311.6442089			411.271935
1396.399	200	311.6274622			411.2085535
1396.449	200	311.6107181			411.1451909
1396.499	200	311.5939765			411.0818473
1396.55	200	311.5772374			411.0185228
1396.6	200	311.5605008			410.9552172
1396.65	200	311.5437667			410.8919307
1396.7	200	311.5270352			410.8286631
1396.749	200	311.5103061			410.7654145
1396.799	200	311.4935796			410.7021849
1396.849	200	311.4768555			410.6389742
1396.899	200	311.460134			410.5757825
1396.95	200	311.443415			410.5126098
1397	200	311.4266985			410.449456
1397.05	200	311.4099845			410.3863212
1397.099	200				

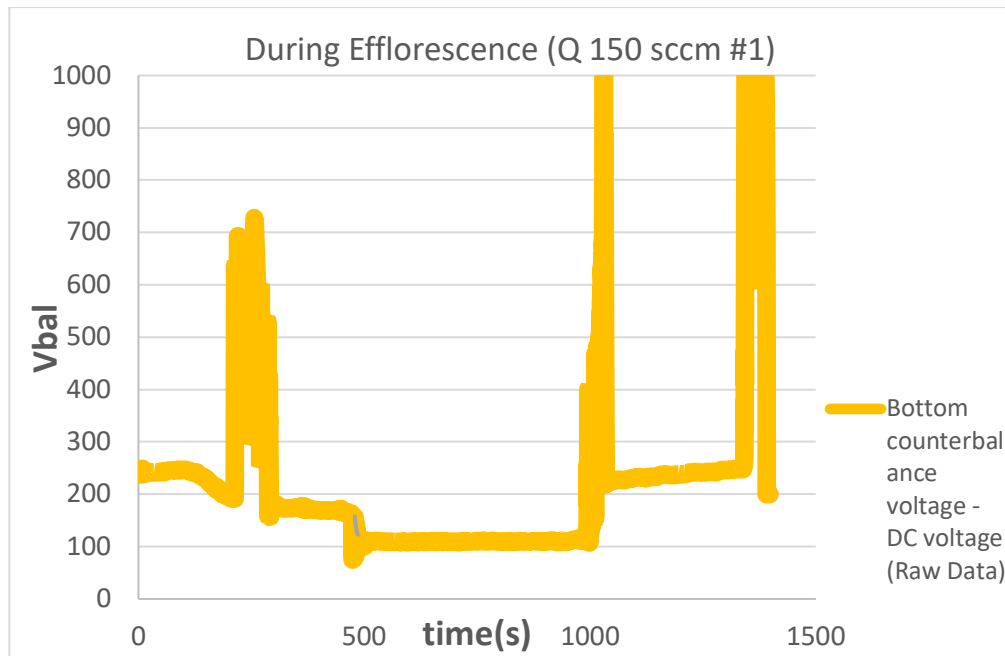
**Table 7:** Data used to analyze the hygroscopicity cycle of NaCl droplet #1 in the EDB device at 150sccm flow rate during the conduction of the NaCl deliquescence/efflorescence experiment (1) With noise signals (2) With minimum noise signals, after efflorescence occurs.

## APPENDIX B

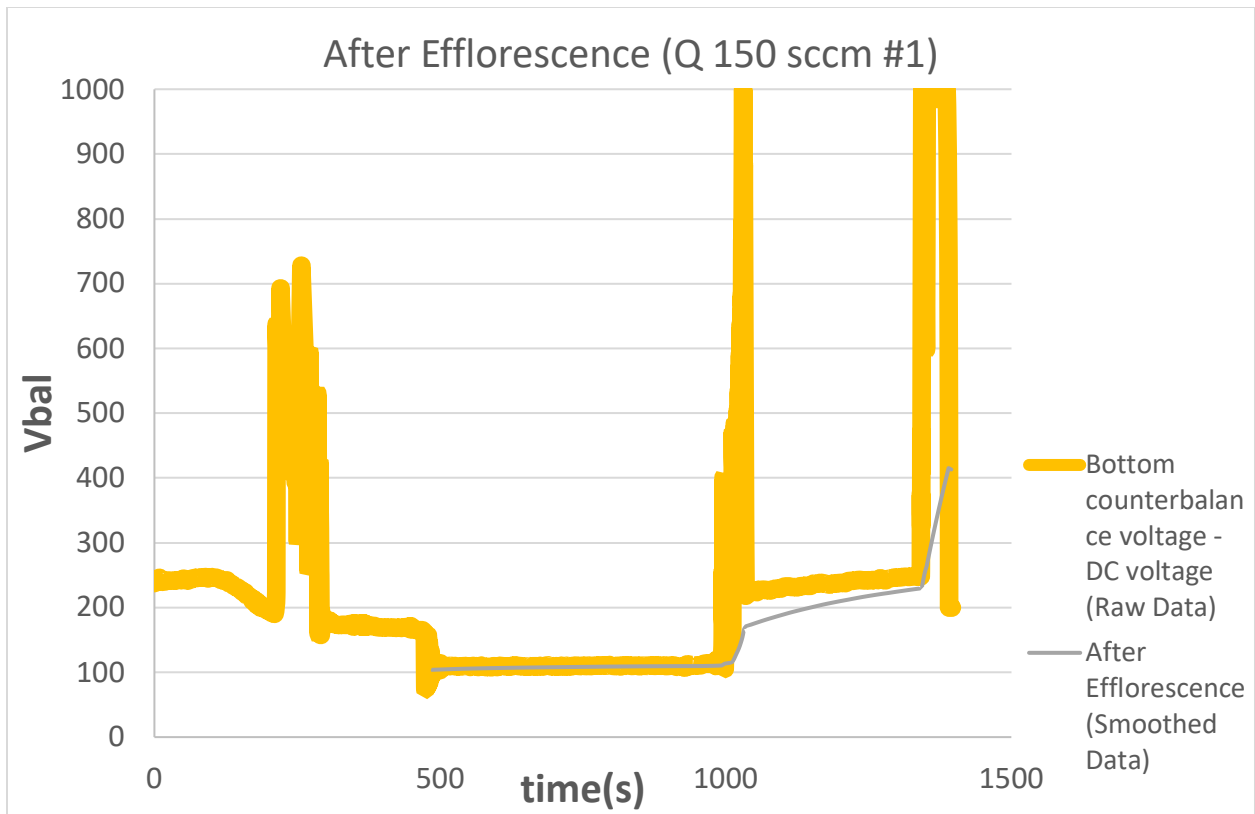
### Data Smoothing: Comparison of hygroscopicity cycles of 5wt% NaCl droplet before, during and after efflorescence



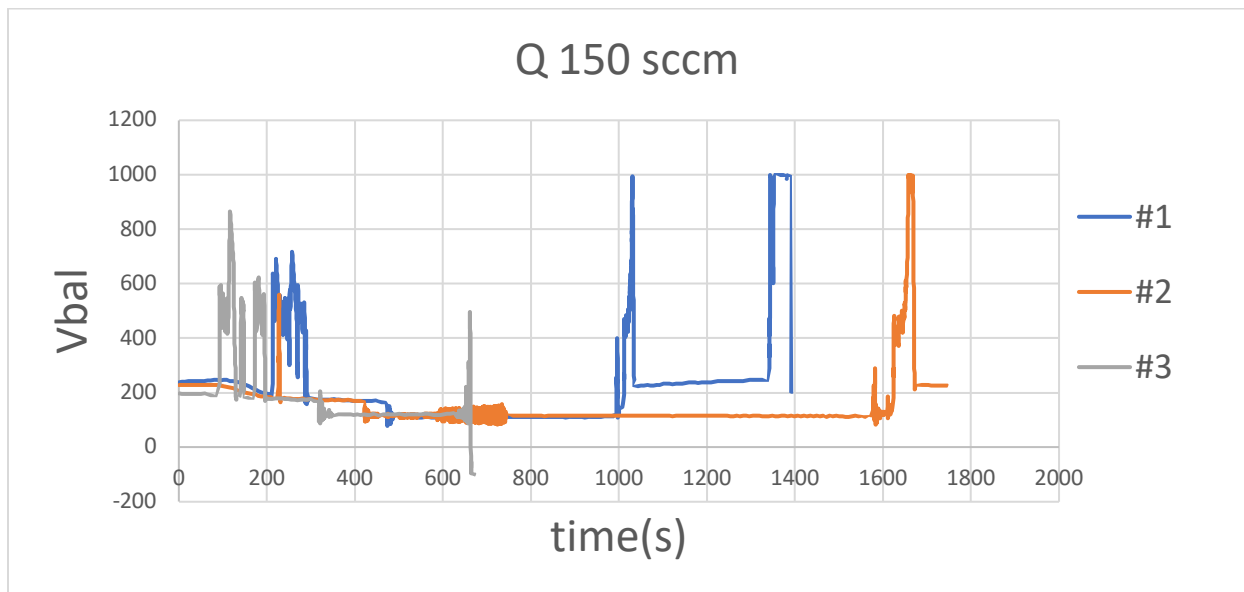
**Figure 10:** Comparing the hygroscopicity cycles of NaCl droplet #1 in the EDB device at a flow rate of 150sccm (1) With noise signals (2) With minimum noise signals, before efflorescence occurs.



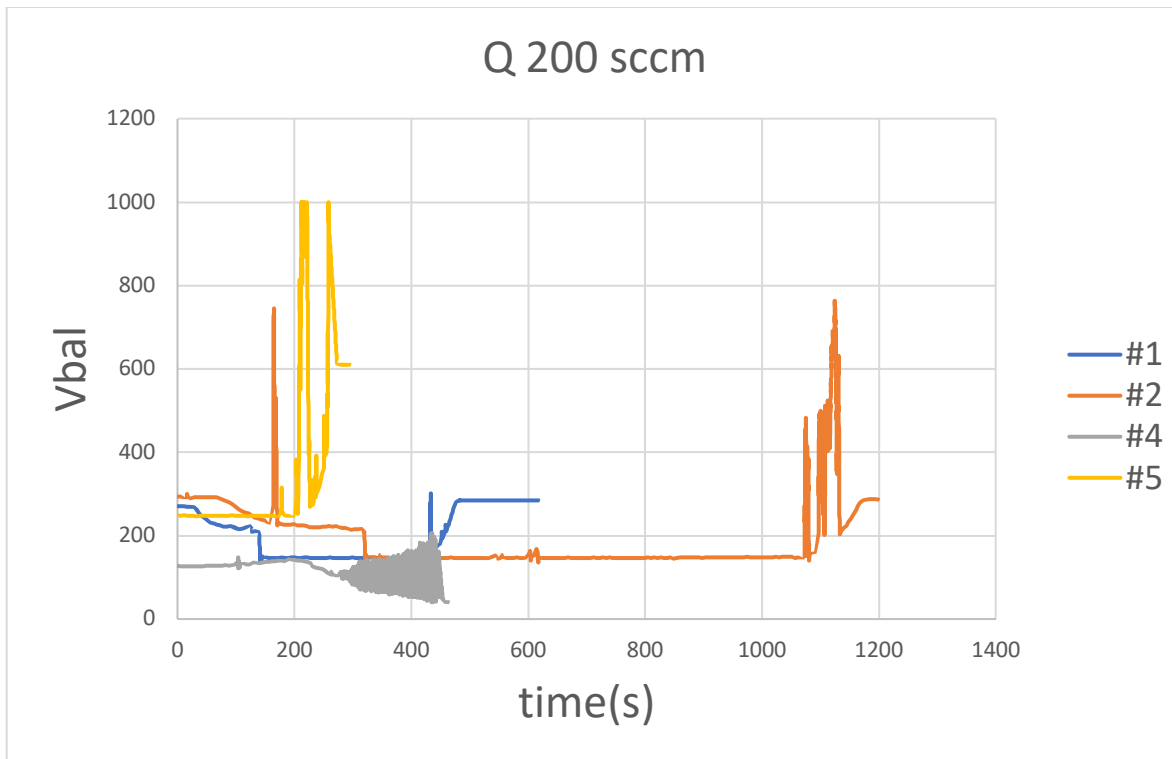
**Figure 11:** Comparing the hygroscopicity cycles of NaCl droplet #1 in the EDB device at a flow rate of 150sccm (1) With noise signals (2) With minimum noise signals, during efflorescence occurs.



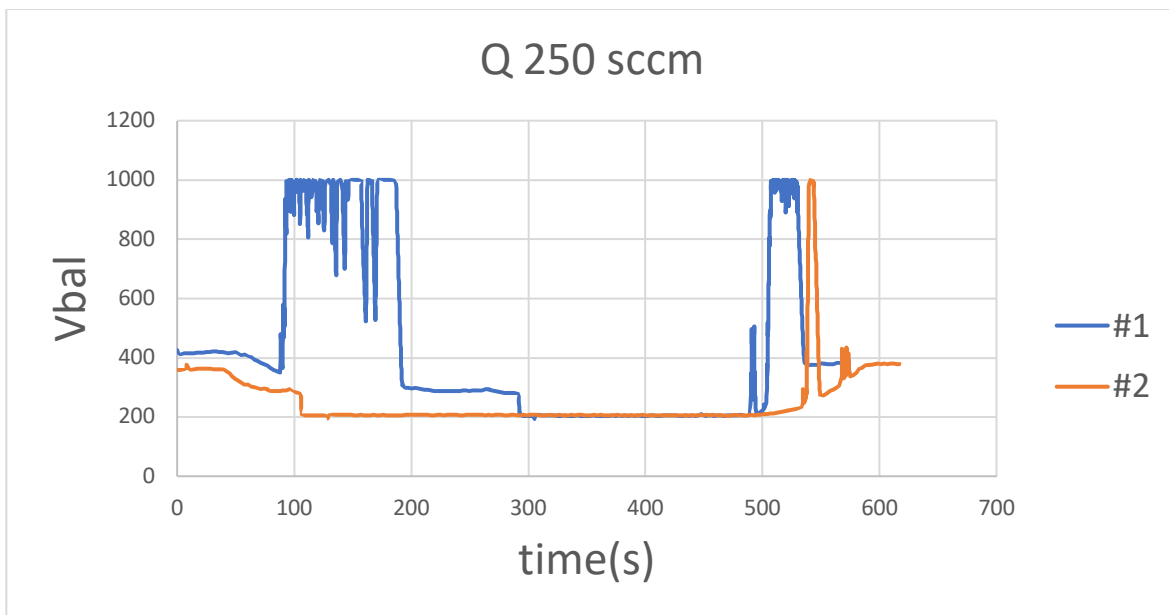
**Figure 12:** Comparing the hygroscopicity cycles of NaCl droplet #1 in the EDB device at a flow rate of 150sccm (1) With noise signals (2) With minimum noise signals, after efflorescence occurs.



**Figure 13:** Hygroscopicity cycle of 3 different NaCl droplets in the EDB device at a flow rate of 150sccm.

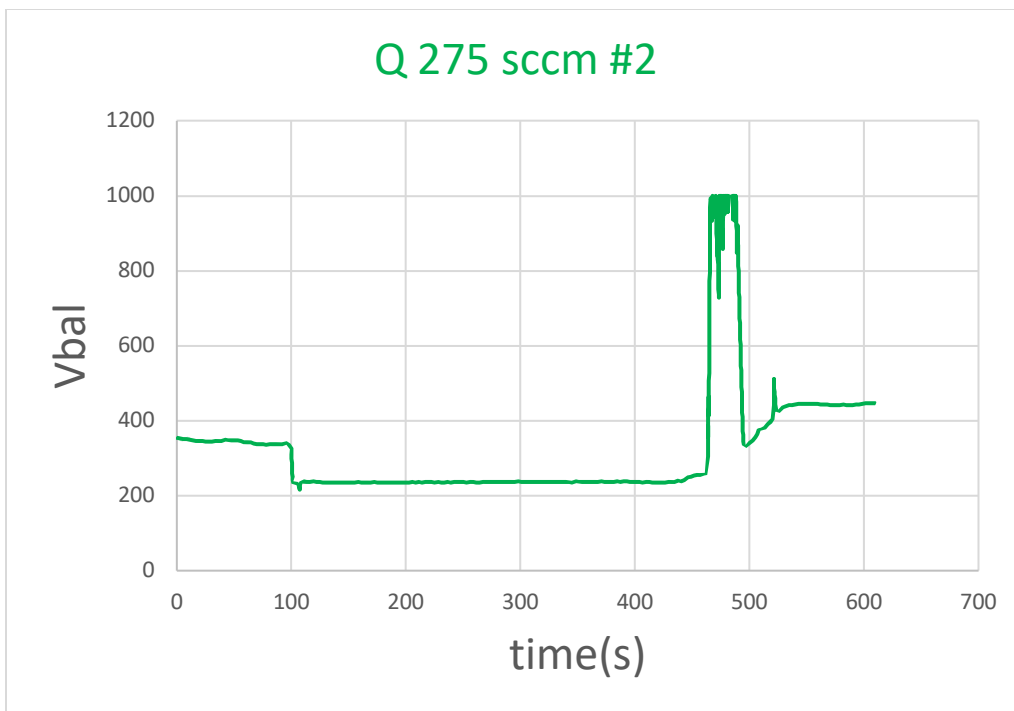


**Figure 14:** Hygroscopicity cycle of 4 different NaCl droplets in the EDB device at a flow rate of 200sccm.

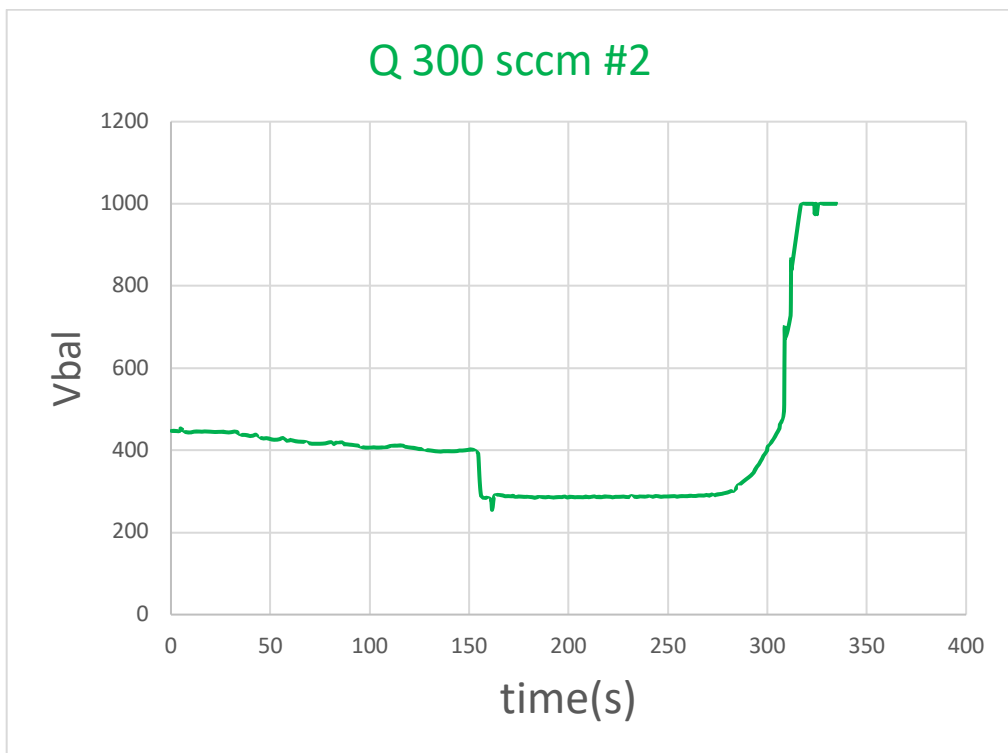


**Figure 15:** Hygroscopicity cycle of 2 different NaCl droplets in the EDB device at a flow rate of 250sccm.





**Figure 16:** Hygroscopicity cycle of a NaCl droplet in the EDB device at a flow rate of 275sccm.



**Figure 17:** Hygroscopicity cycle of a NaCl droplet in the EDB device at a flow rate of 300sccm.

## APPENDIX C

### Definitions

#### Chemical Potential

The chemical potential of a specie in thermodynamics is the energy that can be absorbed or released owing to a change in the particle number of the given species, such as in a chemical reaction or phase transition. The chemical potential of a species in a mixture is defined as the rate of change of a thermodynamic system's free energy with regard to the number of atoms or molecules of the species introduced to the system. As a result, it is the partial derivative of the free energy with respect to the amount of the species, with the concentrations of all other species in the mixture remaining constant.

The chemical potential is the **partial molar Gibbs free energy** when both temperature and pressure are held constant, and the number of particles is represented in moles. The whole sum of the product of chemical potentials and stoichiometric coefficients is zero in chemical equilibrium or in phase equilibrium because the free energy is at a minimum. The chemical potential of any chemical species is universally the same throughout a system in diffusion equilibrium.

The chemical potential drives mass transfer in the same way that the thermal potential drives heat transfer from higher to lower temperatures.

#### Chemical Equilibrium

Chemical equilibrium is the thermodynamic equilibrium in a system that allows for both direct and reverse chemical reactions. If the system achieves chemical equilibrium, the rates of all reactions proceeding in opposite directions are equal. As a result, the system's macroscopic parameters remain constant, and the relationship between the concentrations of reactive components remains constant at any given temperature. In a chemical reaction equation,

equilibrium is expressed by the equality  $\sum v_i \mu_i = 0$ , where  $\mu_i$  is the chemical potential of each reagent ( $i = 1, 2, \dots$ ) and  $v_i$  is the stoichiometric coefficient of each substance (it is positive for initial substances and negative for products of a reaction).

Chemical equilibrium exists when a system's chemical composition does not change. For example, if an arbitrary combination of  $H_2$ ,  $O_2$ , and  $H_2O$  is enclosed in a vessel at a specified temperature and pressure and no future change in chemical composition occurs, the system is in chemical equilibrium. It should be noted that the three species are permitted to react chemically, with the restriction that the number of moles of a species consumed must match the number of moles produced, i.e., there is no net change in the concentration of any species.

## Mole Fraction

The mole fraction or **molar fraction** ( $x_i$  or  $\chi_i$ ) in chemistry is defined as a unit of a constituent's amount (expressed in moles),  $n_i$ , divided by the total amount of all constituents in a mixture (also represented in moles),  $n_{tot}$ . This expression is as follows:

$$x_i = \frac{n_i}{n_{tot}}$$

The sum of all the mole fractions is equals 1:

$$\sum_{i=1}^N n_i = n_{tot}; \sum_{i=1}^N x_i = 1.$$

The mole percent, molar percentage, or molar proportion (mol%) is the same notion represented with a denominator of 100. Amount fraction is another name for the mole fraction. It is the same as the number fraction, which is defined as the number of molecules of a constituent  $N_i$  divided by the total number of molecules  $N_{tot}$ . The mole fraction is a dimensionless quantity that can be used to indicate the composition of a mixture.

## **Raoult's Law**

The vapor pressure of a solvent above a solution, according to Raoult's law, is equal to the vapor pressure of the pure solvent at the same temperature scaled by the mole fraction of the solvent present:

$$P_{solution} = \chi_{solvent} P^o_{solvent}$$

## **Non-Ideal Solution**

A non-ideal solution is one that deviates from the ideal solution's norms, in which the interactions between molecules are the same (or extremely similar) to those between molecules of different components. That is, no Van der Waals nor Coulomb forces, nor any other kind of force, are acting between the parts. We presume that diluted solutions have ideal properties.

## **Thermodynamic Activity**

Activity (symbol  $a$ ) is a measure of a species' "effective concentration" in a mixture according to chemical thermodynamics. This means that the chemical potential of a species depends on the activity of a real solution just as it would depend on concentration for an ideal solution.

Activity is typically thought of as a dimensionless quantity, despite the fact that its value relies on the species' chosen standards of normal behavior. Pure substance activity in condensed phases (solid or liquid) is typically regarded as unity (the number 1). In addition to other factors, activity is influenced by the mixture's composition, pressure, and temperature. The effective partial pressure, also known as fugacity, is the activity for gases.

Because interactions between different types of molecules in non-ideal gases or liquids differ from interactions between the same types of molecules, there is a discrepancy between

activity and other measurements of concentration. The environment has a significant impact on an ion's activity.

## **Molality**

The number of moles of solute in a solution equal to 1 kg or 1000 g of solvent is referred to as its molality. The definition of molarity, on the other hand, is based on a certain volume of solution.

'mol/kg' is a typical molality measurement unit in chemistry. One molal is another way to refer to a solution with a concentration of 1 mol/kg. Molar mass must be represented in *kg/mol* rather than the more common *g/mol* or *kg/kmol* when using the unit *mol/kg*.

The amount of substance (in moles) of the solute,  $n_{solute}$ , divided by the mass (in kg) of the solvent,  $m_{solvent}$ , determines a solution's molality ( $b$ ):

$$b = \frac{n_{solute}}{m_{solvent}}$$

When there are multiple solvents present in a solution, the molality of the mixed solvent can be determined by treating it like a pure pseudo-solvent. Units are defined as mole solute per kilogram mixed solvent rather than mole solute per kilogram solvent as in the binary case.

## **Electrolyte Solutions**

A solution known as an electrolyte is one that is electrically conductive and typically contains ions, which are atoms or molecules that have received or lost electrons. They are frequently referred to as ionic solutions because of this, however there are some situations in which the electrolytes are not ions.

The idea that like charges repel and opposite charges attract is a fundamental tenet of

electrostatics. The electrostatic attraction is likewise very difficult to overcome.

### **Aerosol Liquid Water Content (ALWC)**

To determine the mass of water absorbed by a unit volume of dry aerosol, the relative ALWC is defined as the ratio between ALWC and the dry aerosol volume  $V_{\text{dry}}$ .

The relative ALWC can be used as a stand-in for an aerosol species' hygroscopicity.

### **Chemical Potential of water**

Water's chemical potential is defined as the free energy per mole of water. Simply described, chemical potential is the ability of a substance to react or move, or to conduct work. When a force is applied to an object, it moves from one position to another.

Water potential is the chemical potential of water, and its value is proportional to the quantity of water molecules in the system. The combination of solute potential plus pressure potential yields water potential. The potential for solute is negative, but the potential for pressure is positive.

### **Temperature Dependence of Relative Humidity (RH)**

With the same absolute/specific humidity, air in a cooler environment will have a higher relative humidity than air in a warmer environment because warmer air may contain more water vapor (moisture).

The quantity of water vapor that the air is currently carrying as compared to what it would be holding if it were saturated is known as relative humidity. For instance, if the relative humidity is 20%, the air has 20% of the water vapor it is capable of holding at that temperature. However, as the air's ability to contain water vapor increases with temperature, the relative humidity drops.

## Solubility of Aerosol Salts as a Function of Temperature

### *Effect of temperature on liquid and solid solutes*

Depending on whether the dissolution reaction is exothermic or endothermic, a solid or liquid's solubility may change as temperature rises.

Solubility increases as temperature rises

- In endothermic dissolution reactions, heat energy is absorbed into the system as the solute dissolves due to the net energy from breaking and creating bonds. Additional heat energy is added to the system when the system's temperature rises.
- Le Chatelier's Principle states that the system will respond to this rise in temperature by encouraging the dissolving reaction to absorb the extra heat energy. Solubility of the solute will consequently increase as temperature rises.
- Ammonium nitrate, which is used in first-aid cold packs, is an example of a solute whose solubility rises with increasing temperature. An endothermic process occurs when ammonium nitrate dissolves in solution. As the ammonium nitrate dissolves, heat energy from the surroundings is absorbed, making the area feel cold.

Decreasing solubility as temperature rises

- When a solute dissolves in a solution, heat energy is released in an exothermic reaction. More heat enters the system as the temperature rises. The system will respond to this extra heat energy by limiting the dissolving reaction, in accordance with Le Chatelier's Principle. The solute is therefore less soluble at higher temperatures.
- Calcium hydroxide, which is used as an antacid and to treat chemical burns, is an example of a solute whose solubility declines with temperature.

## **Deliquescence**

A substance collects moisture from the air through the process of deliquescence, which continues until the substance dissolves in the water and produces a solution. Deliquescence happens when the solution's produced vapor pressure is lower than the partial pressure of airborne water vapor.

If the air is sufficiently humid, all soluble salts will dissolve. A substance is referred to as hygroscopic if it can absorb moisture from the air without necessarily dissolving.

## **Efflorescence**

When exposed to air at room temperature, crystalline molecules exhibit efflorescence, which is the loss of water. A chemical substance absorbs moisture from the air and dissolves in it to form a solution through the process of deliquescence.

Efflorescence happens when the hydrate's aqueous vapor pressure is higher than the partial pressure of the air's water vapor.

## **Multicomponent Aerosols**

Only at the eutonic point do particles dried from multicomponent aqueous aerosols have a homogeneous chemical shape, according to thermodynamic study. The particles are made up of a pure salt core surrounded by a mixed salt coating, where the coating's chemical makeup is similar to the eutonic point and is unaffected by the original component mole fractions while the core's composition is exclusively dictated by the original aerosol's composition.<sup>34</sup>