



ISSN: 2350-0328

**International Journal of Advanced Research in Science,
Engineering and Technology**

Vol. 6, Issue 1, January 2019

Experimental investigation of upside down convection and metastable convection cell in water

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ABSTRACT: In this paper we report on the observation of upside down convection cells in columns of partially supercooled water and the conversion of upside down convection cells to metastable convection cells and back to upside down convection cells. During upside down convection a 10cm tall column of water with heat input at the top of 700mW was cooled to below 0°C. As long as the bottom tip of the column of water remains supercooled to several degrees below 0°C and latent heat was not released anywhere in the column the input heat at the top flow down; even when the temperature at the top of the column was above 0°C or raised to far above 0°C. Upside down convection can be imbibed or stopped by the addition of a solute such as heavy water; D₂O. If the bottom of the water column is supercooled and latent heat is not released when the solute is added then a metastable convection cell will develop. It may last for several hours before it dissipates and upside down convection begins.

I. INTRODUCTION

There are numerous research reports of studies of convection currents in water. In all of these cases the water was confined in various shape containers and volume [1, 2]. In most of these studies heat was introduced into the water from the bottom or the side [3, 4, 5]. In some cases ice was involved either; on top of the water or at the bottom of the water column [6, 7]. In this study we did not introduce heat in order to induct convection; rather we removed heat by cooling from the bottom. By cooling to below 0°C we induce what we call “upside down convection” (UDC). Upside down convection means warm water flowing down and colder water rising up a column of water. In 1968, George Veronis wrote, “A stabilizing gradient of solute inhibits the onset of convection in a fluid which is subjected to an adverse temperature gradient. Furthermore, the onset of instability may occur as an oscillatory in temperature because of the stabilizing effect of the solute on convection” [8]. In Veronis’ case of both theoretical and experimental studies, heating was from below. Here we cooled from below. The primary solute used was heavy water (D₂O). However, most inorganic and organic salts will work if they are dissolved in water before adding to the test water. In this paper we show and discuss the onset of UDC and several examples of metastable thermal rotating convection cells. The best convection cells were produced in partially supercooled water where the top of the columns were near or above ambient temperature and the bottom was supercooled to several degrees below 0°C. The bottom of the water columns must be below 4°C and preferably supercooled and may or may not have a solute added before cooling. If during an experimental run latent heat is released, (UDC) and/or thermal oscillations will be abruptly ended because the bottom of the column of water will no longer be supercooled; it will now be at 0°C and in the process of freezing.

Upside down convection begins as soon as the temperature of the water at the bottom of the column falls below 4°C. Between 4°C and 0°C UDC is weak; however, it increases dramatically as the water supercools (See Fig. 2). Under these conditions hot water moves down the column and cold water moves up the column. This process will cool the whole column to 0°C or lower even when there is heat input at the top (See Fig. 2). The oscillations often appear near the bottom of the column as a rotating cell, if a solute has been added and has not yet homogeneously mixed with the pure water (See Fig. 7). When a solute was added after the onset of UDC, the onset of UDC was abruptly ended for a



ISSN: 2350-0328

International Journal of Advanced Research in Science, Engineering and Technology

Vol. 6, Issue 1, January 2019

period of time determined by the solute used and the volume of solute added. The time of inhibited upside down convection may vary from minutes to hours.

This paper reports on the methods and data that demonstrates the occurrence of the Upside Down Convection phenomena, but more sophisticated equipment would be useful in providing further evidence.

II. LITERATURE SURVEY

Studying convection of water that is purposefully cooled from the bottom is not as common as studying convection when the water is heated from the bottom. A popular type of water convection that is demonstrated in nature, particularly in ocean waters, is penetrative convection. Penetrative convection occurs from updrafts or thermals that can reach the top stable fluid layer and translocate back to the bottom fluid layer [9]. A prime proponent of penetrative convection is capping inversion, where an elevated fluid layer is positioned above the boundary layer interacting with the surface below it. In simulated research experiments, that surface interacting with the boundary layer would be some type of container, test tube, or other holding object. Thermals, when warmer air rises and crashes into the colder air due to the density decrease, help create the capping inversion. The displacement of the warm & cold air relationship is executed where the lighter-warm air now operates at the top and the denser-cold air now operates at the bottom [10].

In 1910, Birge proposed and indicated the effect of solar heating the submerged ice temperature water causes some amount of convection with temperature configurations. Specifically Birge mentioned that temperature about 4 degrees Celsius can make convection currents can transfer heat from the ice submerged water. Birge's proposal is further supported by mid to late 19th century photographs taken and submitted by multiple researchers detailing cell-like patterning relative to Rayleigh-Benard convection [11]. Earlier photos taken during the very early 20th century detailed the same patterning but had remnants of solar radiation that penetrating the ice layer causing heat flux to spear point of the convection itself followed by measurements of an aspect ratio of the observed convection cells comparing width of the convection cell to the location depth of the convection cell [11].

To the field of science, convection is known as the transfer of heat from the sum fluidic molecular movement in the phases of matter such as gases and liquids. Thermal convection can be produced using a heat source to send heat on the sides of an object that is filled with liquid, and we should all see changes in temperature of the liquid inside the glass due to warmer fluid circulating into the cooler areas. Convection, also known as convective heat transfer, is a fundamental type of heat transfer and it takes places by the random Brownian motion of fluidic individual particles and by advection, where matter or heat is transported by greater motions of the liquid (Townsend 1964). Many endeavors in the study of the convection of water have a frequent and specific methodology or experimental setup. The frequent and specific setup is (1) an ample amount of liquid, mainly distilled water, inside an apparatus (completely closed or opened at the top) of a cylindrical or rectangular shape; (2) a heating source under the bottom or on the sides of the apparatus that provide the stimuli to produce convection; and (3) a type of temperature measurement device or construction to help gather data on the rate and duration of the changes in the temperature of the liquid versus the surrounding environment. Over decades there have been many other innovative experimental studies regarding the convection of water with unique designs to try to mimic convection occurring in different environments and situations. Those experiments used water or fluid that was already cold or placed a fluid-filled apparatus in a body of cold water to study the convection of cold water.

The Townsend study was conducted to observe and better understand how convection was produced from disturbing the layer of stably-stratified fluid from an ice-covered bottom [1]. The convection within the underlying layer of unstable fluid led to temperature fluctuations caused by impactful overturning of the water particles inside the water tank. The water molecules that were confined to the thin layer of ice-covered bottom displayed floating columns in rising fluid [1]. Other observable physical and spatial characteristics of the water molecules included the floating rising water columns penetrating a region that maintained a constant average temperature that was 3.2°C and that occupied at least two-thirds of the water tank volume [1]. The water columns in the tank were reported to be horizontally stretching or at least positioned to reach the measured region of stable stratification [1].

In 2003, the group of researchers from the Instituto Tecnológico de Veracruz and the Facultad de Ingeniería in Mexico in 2003, conducted research on water's natural convection coming close to its density inversion [6]. A square-shaped cavity apparatus was used with two detailed adiabatic side walls originally lining up vertically and the apparatus contained already cold water that was put into the density inversion range. The heat source came from the bottom of the square cavity to produce convection with a uniform continual temperature to be compared with the cavity's top surface temperature [6]. The square cavity apparatus was not tilting or moved at any time during the research experiment, but the adiabatic side walls were tilted. Natural convection cells were produced even with the tilted angle of the adiabatic side walls due to the vertical direction of the square cavity. The tilted angle affected the size and shape dimensions of the natural convection cells [6]. The results of that study were a stark contrast to the base results of the research team of Inaba and Fukuda [7] that observed greater unicellular flow and the highest temperature gradients being the closest to the isothermal walls [7].

III. METHODOLOGY

A method for producing UDC in a column of water is schematically shown in Fig. 1. Upside down convection was most dramatic when the outer walls of the tube holding the the water was insulated from ambient air. When this was the case, heat input was restricted to the top of the column and must flow all the way down the column. A well-insulated tube will produce dramatic results as shown in Fig. 2

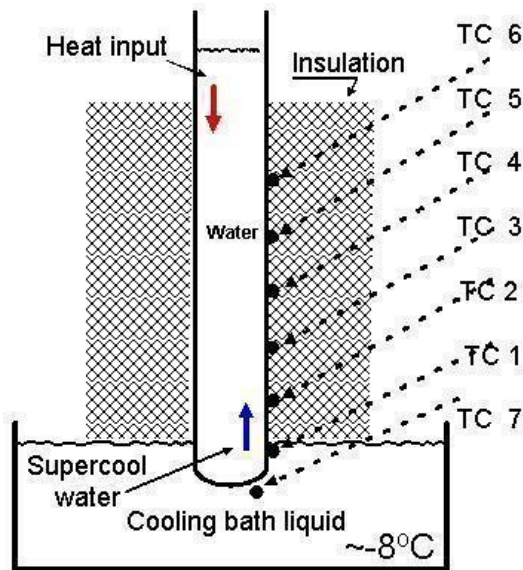


Fig. 1. A typical experimental set-up is a water filled tube 10cm long and cm in diameter. Thermocouple 7 is inserted into the cooling bath in order to measure the temperature of the cooling bath. Thermocouple 8 is moving up and down the inside of the water tube in order to obtain accurate measurements of the temperature changes of the water. The top of the tube is open and is exposed to ambient air or a heat source. D₂O or other solutes can be added at the open top.

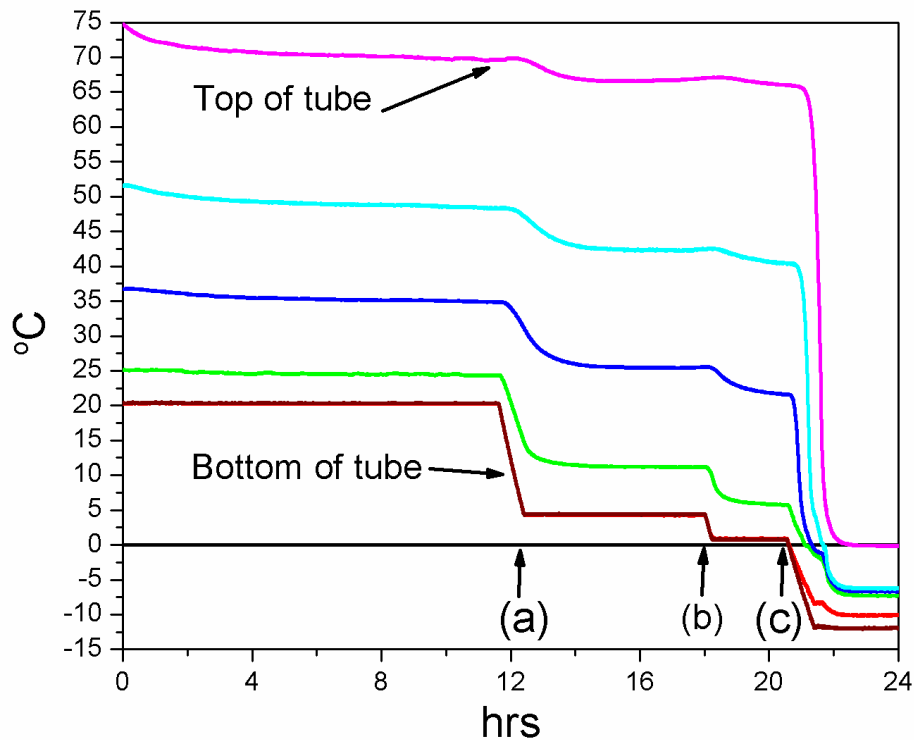


Fig. 2. Here we show an example of the dramatic onset of upside down convection in pure water (no added solute), when the temperature of the water at the bottom of the tube was lowered to below -10°C as our experimental control. At which time the water at the top of the column, which had been held at $>65^{\circ}\text{C}$ for more than 20 hours, suddenly fell to 0°C . In this setup, we only used 7 thermocouples to measure the temperature instead of 8 thermocouples.

Type K (Ga. 0.010) thermocouples (TC) were attached to the outer walls of glass test tubes with epoxy as illustrated in Fig. 1, a typical tube was glass, 10cm long and $\sim 1\text{cm}$ in diameter. It was encased in a Styrofoam insulating carriage that was $\sim 6\text{cm}$ thick with the option for an open or closed top with the bottom of the tube exposed. The exposed part at the bottom was about 1cm long and was inserted into the cooling bath. The top was otherwise exposed to the ambient environment or a heat source.

To produce metastable rotating convection cells a solute was added to the column of water. It may be added at any time, however, it must be added gently as not to cause homogenous mixing. This can be done with a pipette or droppers held a few millimeters above the surface of the water. The data acquisition system consists of an eight channel Omega Engineering module for thermocouples, the OM-DAQ-USB-2400.

To begin an experimental run the bottom of the tube was placed in the cooling bath as shown in Fig. 1. It was then filled with water and the chiller was turned on. The chiller in this case was a Lauda RM 6 Chiller. However, any cooling system that will cool a cooling bath to -4 degrees Celsius or lower will work. The important point here is to supercool the water as low as possible before latent heat is released. If at any time latent heat is released and freezing begins UDC may either; not begin or it will be terminated instantly if it has begun, see Fig. 3. If latent heat is released fresh water should be added and the cooling bath temperature raised slightly. If freezing becomes a problem keep raising the cooling bath temperature until the water remains supercooled for hours or even days. Once that temperature has been determined experimentation can begin.

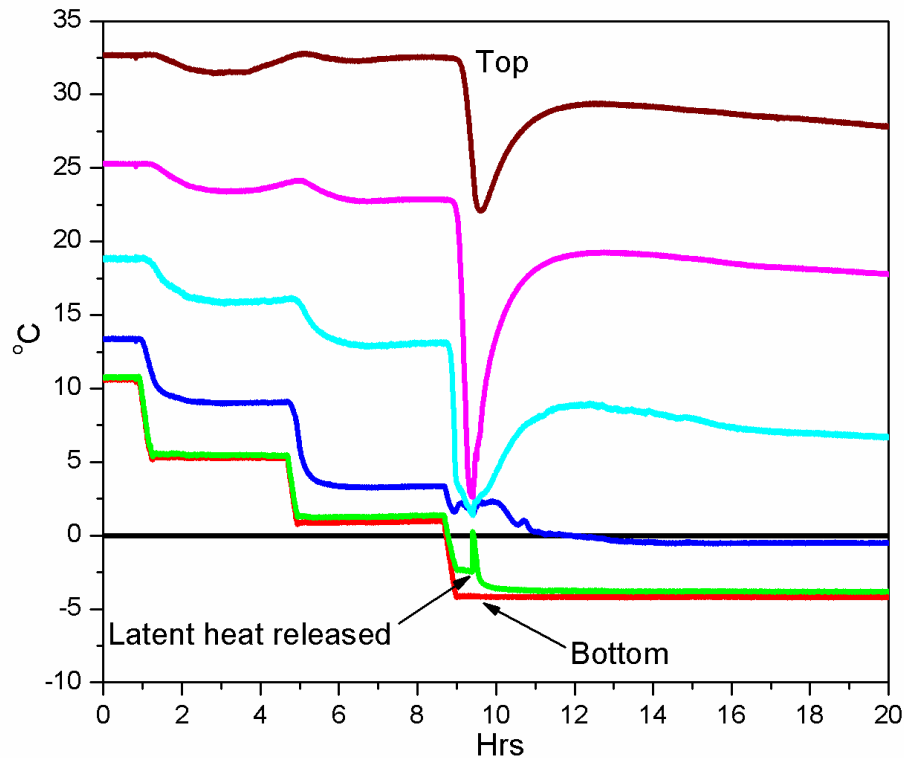


Fig. 3. At ~9 hours into the run the temperature of the water at the bottom of the column was supercooled to -4°C and UDC began, the temperature of the water at the top of the column began to fall. In this setup we used 7 thermocouples as evidenced by the 7 colored curves. About 30 min. later latent heat was released and UDC ended, at that time the temperature of the water in the upper part of the tube began to rise while the water at the bottom continued cooling and froze to solid ice. In this setup there is constant heat at the top of the column. When UDC stops, heat is taken out or reduced at the bottom of the water column, causing the temperature at the top of the column to rise. The temperature at the top of the column will to the ambient temperature level.

III. RESULTS AND DISCUSSION

Data presented in Fig. 2 was collected using a tube configured as shown in Fig. 1. In this case seven thermocouples were used to record the temperature in and around the tube while the eighth was in an ice/water bath, a 0°C reference. This allowed us to correct for any electronic drift during long runs, up to several days.

For the data shown in Fig. 2 the system was held with a thermal gradient of $\sim 50^{\circ}\text{C}$ from top to bottom for ~ 12 hours. After 12 hours the temperature at the bottom of the column was lowered to $\sim -4^{\circ}\text{C}$ at (a) in Fig. 2 with minimum effect on the temperature at the top of the column. At (b) the temperature was further lowered to just above 0°C , again with minimum effect on the temperature at the top of the column. Then at (c) the temperature of the water at the bottom of the column was lowered to $< -10^{\circ}\text{C}$ then, suddenly temperature of the water at the top fell to $\sim 0^{\circ}\text{C}$. Heat was now flowing down the column from the heater ($\sim 700\text{mW}$) at the top. This was UDC, cold water (-10°C) flowing up and hot water (65°C) flowing down. If latent heat is released any time after (c) in Fig. 2 the temperature of the water at the bottom of the column will rise to 0°C and the temperature at the top will return to $> 65^{\circ}\text{C}$. See for example Fig. 3.

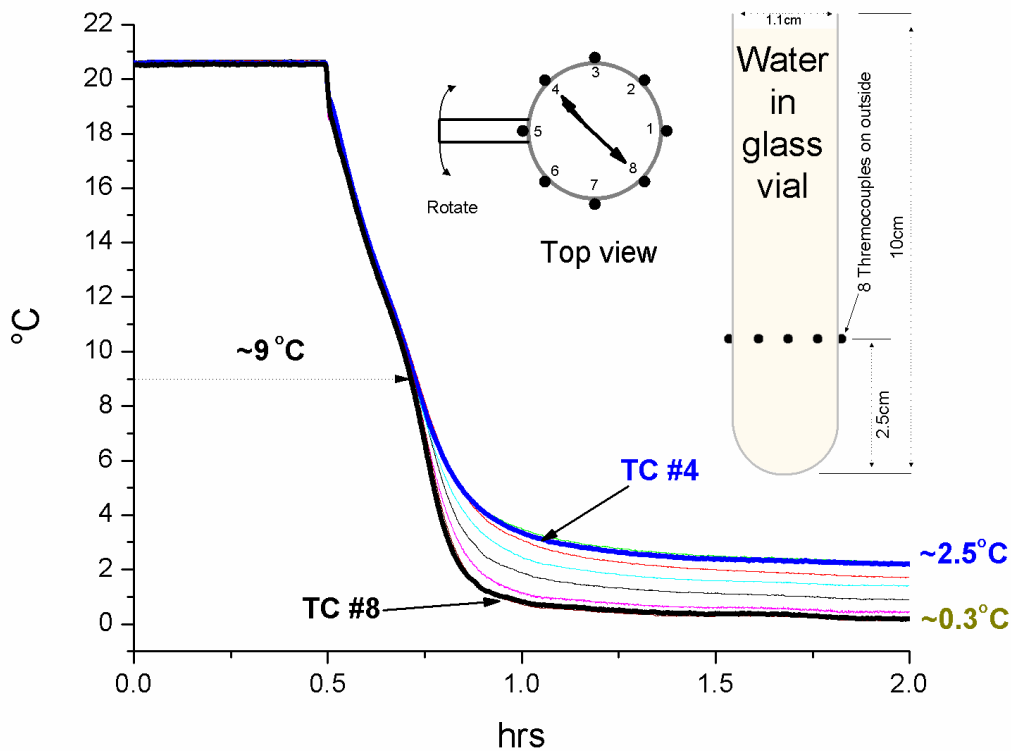


Fig. 4. The insert shows an alternate set-up with the thermocouples on the outside of the tube 2.5cm from the bottom. The thermocouples are spaced at 45° apart around the outside of a tube. The tube can be manually rotated 180° around its vertical axes and/or tilted from its vertical axis by 15° in any direction.

The temperature traces shown in Fig. 4 are of all eight thermocouples as the water is cooled from the bottom. Notice that after the temperature of the water at the location of the thermocouples falls to ~9°C, a thermal gradient develops across the tube. This is a signature of UDC. This is a stable gradient of more than 2°C over a horizontal distance of 1cm. The stability is demonstrated in Fig. 5 which is a later part of Fig. 4. This shows what happens when we manually rotated the tube 180° around its vertical axis 3.8 hours into the run. The columns of cool and warmer water within the column of water remains fixed in their vertical position as the tube holding the water was rotated 180°.

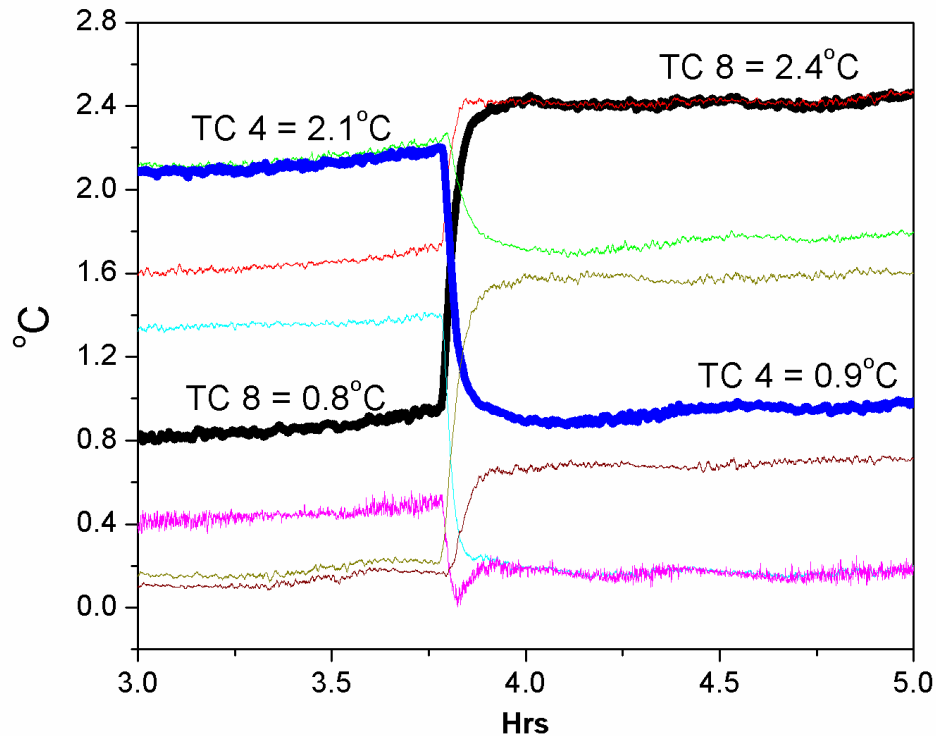


Fig. 5 Here we show an expanded, zoomed-in region of Fig. 3 beginning 3 hours into the run. At about 3.8 hours the tube was rotated 180° around its vertical axis. The set-up is as shown in the inset in Fig. 4. Notice the dramatic change in the temperature between thermocouples 4 and 8 after the rotation.

Using the configuration shown in the inset in Fig. 4 we were able to observe many of the phenomena that can be produced during UDC when we make a change in geometry, chemistry or temperature of the water. In Fig. 5 we show the effect of manually rotating the tube 180° around its vertical axis. The vertical convection cells that were produced in Fig. 4 at the onset of UDC in the water are at different temperatures because the vertical convection cells are stable and independent of the horizontal rotation of the tube. This is clearly shown in Fig. 5 when we manually rotated the tube 180° and the temperature that two of thermocouples (TCs 4 & 8) registered changes of more than 1°C. The vertical convection cell phenomena were not significantly affected by the manual horizontal rotation of the test tube.

The vertical convection cells are well defined in the columns of water moving up and down within the water, each at a different temperature. These cells remain vertical even when the tube was rotated or tilted from its vertical position. This is clearly shown in Fig. 6 where the temperature of TCs 2 and 4 show little or no change as the tube was tilted 15° towards TCs 1 and 3 back and forth. While the temperature recorded by TCs 1 and 3 showed a change of more than 2°C. This demonstrates that the convection cells adjacent to TCs 1 and 3 shifted from one side of the tube to the other with each tilt of 15° back and forth.

The temperature of vertical convection cells measured by TCs 2 and 4 remain stable as long as the bottom tip of the column of water remains supercooled.

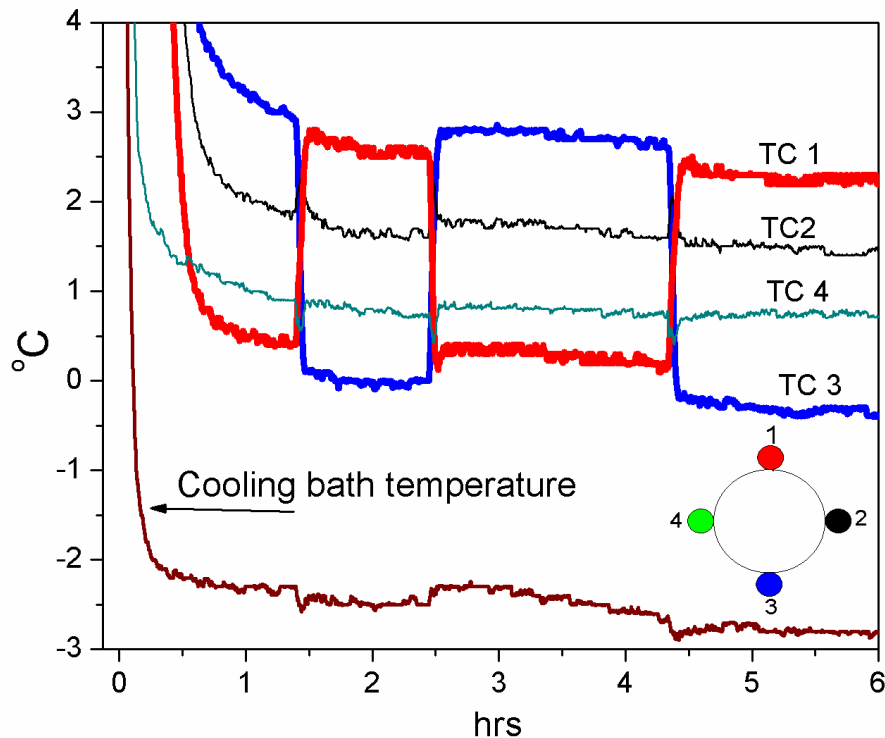


Fig. 6. Here we measured the response of thermocouples 1 and 3 to a tilt of 15° along a line through TCs 1 and 3, which is the vertical axis of the tube in this experimental setup. The tilt was back and forth along the vertical axes of the tube. Note the stability of the horizontal temperature gradient before and after the tilt and almost no change in the temperature of TCs 2 and 4.

This stability in temperature was independent of the number of times the tube was tilted back and forth.

When a solute was added and not homogeneously mixed to a column of water before or even after it had transitioned to a state of UDC, a metastable rotation cell was produced. The cell was located in water where the temperature is below 4°C . A typical example is shown in Fig. 7. Here the solute was D_2O and it was added to the top of the column of water before the start of cooling. In this case $\sim 100\ \mu\text{L}$ of D_2O was added to $\sim 10\ \text{mL}$ of tap water. The tip of the pipette was held about 2 mm above the surface of the water. This insured that mixing did not occur. If homogenous mixing does occur then you will see only vertical UDC. In order to produce rotation convection cells there must be few water clusters that have a much higher concentration of molecules of the solute distributed through the upper part of the water column. Once these clusters start breaking apart and reform, the solute becomes more homogeneously mixed and all water clusters now have the approximately same percentage of D_2O in them and UDC resumes. This is shown at (c) in Fig. 7 at about eight hours into the run when the temperature at the top of the column of water begins to fall.

During the first ~ 6 hours the rate of rotation had slowly decreased from one rotation in 110 seconds at (a) the start of rotation to finishing with a rotation rate of one rotation in 320 seconds at (b) near the end of rotation. As the rotating cell began to break apart, see (b) in Fig.7, UDC that was inhibited by the solute now resumes and the temperature at the top of the column began to fall, see (c) in Fig. 7.

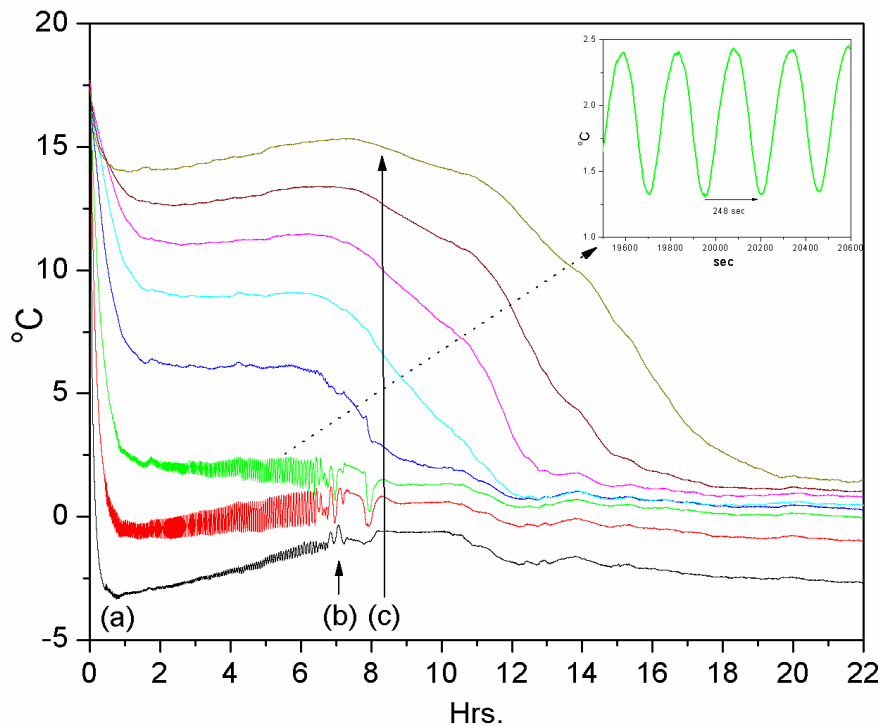


Fig. 7. Here we show the results when a solute (D_2O) was added before the start of cool down. The set-up shown here in Fig. 7 corresponds exactly as the standard setup in Fig. 1. (a) Temperature oscillations start to occur. (b) Temperature oscillations end. Notice that the addition $100\mu L$ of D_2O inhibited upside down convection for about 9 hours starting at the beginning time 0 hours. It took that long for the D_2O to homogeneously mix and acclimate with the water. (c) At which time upside down convection began and the temperature of the water at the top of the tube started falling. There are possible smaller oscillations that were not detected due to the placement of the thermocouples.

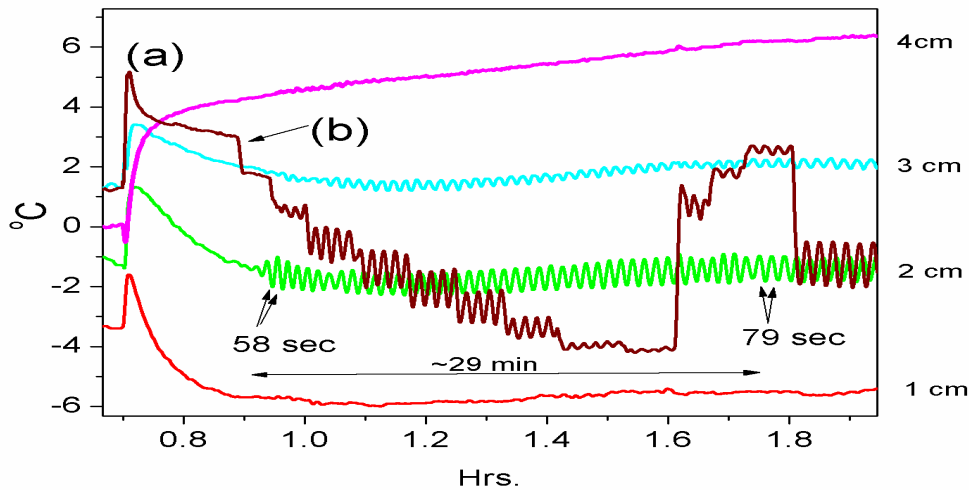


Fig. 8. The vertical width of the rotating convection cell was determined by lowering a thermocouple down the center of a tube holding a column of water, which is represented by the brown trace line. The column of water was in a state of upside down convection before the solute (D_2O) was added at (a). At (b) we began lowering a thermocouple down the tube in millimeter steps of 1-2 mm (approximately 9 total steps for this experimental run). All other thermocouples were fixed 1 cm apart on the outside of the tube beginning 1 cm from the bottom of the tube. The vertical width of the rotating convection cell was between 1.5 - 1.7 cm. In 28 minutes the rate of rotation during this run decreases from once every 58 seconds to once every 79 seconds.

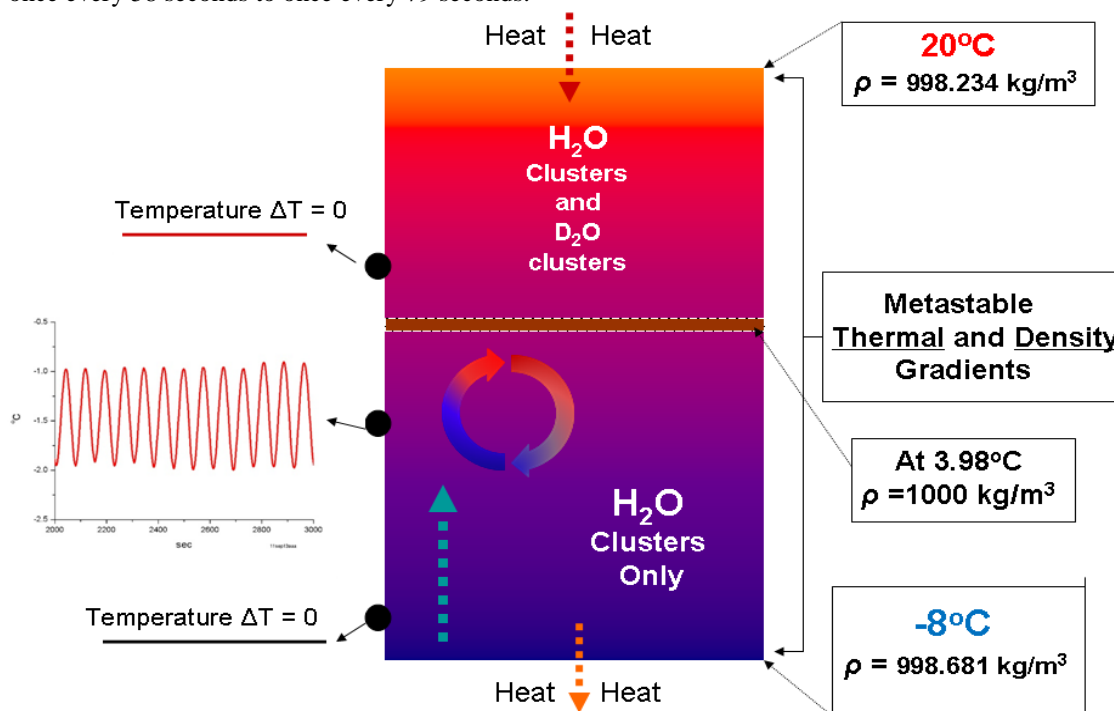


Fig. 9. In Fig.1 we show a schematic diagram of heat flowing down and supercool water flowing up. This resulted in the whole column of water cooling to $0^{\circ}C$ or below after a period of time. Here in Fig. 9 we show schematically how the same system behaves when D_2O was added at the top of the column and not immediately homogeneously mixed. Homogeneous mixing began at the bottom in the supercool water.



In a column of water where heat input is predominantly from the top and cooling is at the bottom a thermal and density gradient was established as depicted in Fig. 9. When a solute was added at the top of the column of water and not immediately homogeneously mixed, convection was inhibited in that part of the column where the solute remained unmixed. If the water at the bottom of the column was supercooling when the D_2O was added, then mixing at the bottom occurs almost immediately resulting in UDC in that part of the column. This is why the convection cells always form at or near the bottom of the column as shown in Fig. 7. If there is not sufficient heat input at the top to maintain a stable thermal gradient then the cell will slowly move up the column as the top of the column cools down.

In Fig. 9 that location is at maximum density where the temperature was $3.98^\circ C$. In water where the temperature was above $3.98^\circ C$ a small percentage of all water clusters have high concentration of the added D_2O . All water clusters below $3.98^\circ C$ have about the same percentage of added D_2O in each cluster because the D_2O has been homogeneously mixed as a result of forces produced by UDC which originated at the bottom of the column in the supercooled water. If the water was not supercooled then the UDC forces are not strong enough to cause homogeneously mixing. In Fig. 2, (c), we show where the force of UDC pushed below $0^\circ C$ water up to the top of the column.

When a well-defined and stable thermal gradient and two opposing density gradients were established as depicted in Fig. 9, UDC was restricted to the lower part of the column. Under these conditions the cold water rises and approaches the layer of water at maximum density, its density increases and it began to flow back down the column. As it flows down the column its density decreases and it begins rise up the column. This sets up a metastable convection cell. The stability and size of this convection cell is illustrated in Figs. 8 and 9.

This effect produces an implied circle pattern shown in Fig. 9 and the oscillation in temperature at the location of TC 2. Notice that TCs 1 and 3 does not show any change in temperature during this time. In time the water clusters above the maximum density line will be broken apart by the force of the upward moving cold water and the rotating convection cell. After all D_2O dominated water clusters are broken apart UDC will extend to the top of the column and the whole column will cool to $0^\circ C$ or below as shown in Fig. 2. If at any time during a run latent heat is released the process will abruptly end and the column of water will begin freezing from the bottom.

IV. CONCLUSIONS AND FUTURE WORK

In this report we present new observations of upside down convection and metastable convection cells in columns of partially supercool water. We show how upside down convection can be stopped and/or inhibited by the addition of a solute; the solute that we used was heavy water (D_2O). However, any solute with a density greater than pure water may be used. For example saturated NaCl or $MgSO_4$ water can be used as the solute. The convection cells are stable enough for the tube to be tilted from side to side thereby forcing the cells to switch from one side of the tube to the other and still remain in tack as discrete vertical columns of water in a larger column of water. The convection cells are equally stable for long periods of time; hours. We have observed the rotation rates of the convection cells that range from about once per minute to as slow as once in about six minutes. The maximum diameter of the cells was 1 cm with the temperature gradient across the cell of more than $2^\circ C$. By using more sophisticated equipment and research methods we can further demonstrate the specifics of the UDC phenomena in supercool water.

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ISSN: 2350-0328

**International Journal of Advanced Research in Science,
Engineering and Technology**

Vol. 6, Issue 1, January 2019

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