# Effect of condensation on surface contact angle

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# Abstract

Understanding the effect of condensation on surface contact angle is important in many applications, e.g., waste-heat recovery systems. This work aims to fill the two knowledge gaps in the literature related to the effect of condensation on the contact angle: (1) how the drop contact angle varies over time during condensation process; and (2) whether the evaporation of the condensed drops can be the reason for the observed contact angle under condensation environment. In this study, three surfaces, polystyrene, Teflon AF, and superhydrophobic surfaces, were used in experiments. The experimental results show that the contact angle between water and all three surfaces decreases with the time when the surface was exposed to the condensation environment. Such change of contact angle over time is caused by the evaporation of the condensed drop, which prompts the formation of a film formed around the condensed drop. The film formed due to the adsorption of water vapor onto the surface, causes a decrease in the contact angle observed over time. A theoretical model which considers the evaporation effect was developed to help to understand the experimental observation. Finally, to demonstrate the importance of the change in contact angle in condensation environment over time, we show its effect on shedding of drops in the context of drop-wise condensation.

### **1. Introduction**

Condensation is a process where water changes from vapor to liquid state. Initially, the condensation is dropwise, occurring during the nucleation, growth, and coalescence of discrete droplets [1]. After time, if there is no departure of the droplets, the continuous coalescence of droplets leads to a film that covers the surface, and the condensation will become filmwise. Dropwise condensation processes are involved in many applications, e.g., waste-heat recovery systems, thermal power plants, chemical refining, and refrigeration. It has been shown that filmwise condensation can significantly increase the thermal resistance of the system causing a low heat transfer rate. To achieve a high heat transfer performance, filmwise condensation is typically avoided by removing the drops (shedding drops) from surfaces [1, 2].

To remove a drop generated due to condensation, an extra stimulus is needed to change the force balance of the drop sitting on a surface. The drop evolves its shape to respond to the stimulus, before drop motion on the surface. Depending on the application, shedding of dropwise condensation can be achieved with the aid of gravity [3], use of surfaces with wettability gradient [4-7], and through the coalescence-induced spontaneous motion of the droplets [8-12]. The use of an electric field to promote dropwise condensation has also been explored [13-16].

Regardless of the methods used for shedding liquid, the drop adhesion force between the drop and surface must be overcome [17]. The surface wettability (i.e., contact angle and contact angle hysteresis) is the critical parameter deciding the adhesion force, affecting the drop shedding process. For example, for shedding droplets from a solid surface using an airflow, it has been suggested that the critical airflow velocity required for the shedding of a droplet is the result of the competing adhesion and aerodynamic forces acting on the droplet. The contact angle can affect both the aerodynamic force and the adhesion force; hence it is the critical parameter governing the shedding process.

To our knowledge, most of the drop shedding-related literature used the contact angle value measured in a laboratory environment (dry condition) [18, 19]. Such a procedure is appropriate for removing the drops which are not generated due to condensation. The value of contact angle can vary when condensation occurs. When condensation occurs, either the air temperature is lowered to its dew point or it becomes saturated with water vapor, so that it cannot hold any more water.

As shown in the literature, the change of either the temperature or relative humidity (RH) can cause the contact angle to change [20-28]. Generally, the contact angle value decreases with the increase of the temperature [20-23]. For example, Adamson developed a model for the temperature dependence of contact angle which relates wetting with molecular adsorption [19]. Adamson's model predicts that the slope of decrease in contact angle with temperature increases with temperature, as observed in [20,21].

The change of the relative humidity can also cause a change of the contact angle. It has been shown that the adsorption of water onto the solid surface can change when RH changes. Since the adsorption isotherm for an absorbent depends on pressure, a change in relative humidity, which affects the partial pressure of water vapor will affect adsorption. The change of adsorption will further change the solid-liquid interfacial tension, hence causing a change of contact angle. For example, metals such as copper [20], and minerals with high surface free energy can have their previously high energy reduced by adsorption of water, leading to an increase in contact angle [27, 28].

Since the condensation process is always associated with the variation of humidity and temperature, the contact angle value under the condensation environment will be different from dry conditions. Yin et al. [29] provided data for contact angles and sliding angles of superhydrophobic surfaces with different low surface energy coatings. The wettability was explored as a function of the environmental parameters to investigate the stability of superhydrophobicity against condensation. They showed combined effects of temperatures between -10 °C and 30 °C and relative humidity between 30% and 90%. The contact angles were reported to decrease with increasing relative humidity and decreasing temperature. However, the work was limited to the stability of superhydrophobic surfaces.

He et al. [30] studied the evolution of the static contact angles as the surface temperature approaches and descends below the dew point. Their study was conducted on a superhydrophobic silicon wafer with PDMS microstructures. They reported the static contact angle between 2-20 °C, with an RH of 30% and ambient temperature of 20 °C. They explain their results using the Cassie equation, and molecular adsorption. In the presence of adsorption, they suggest the solid phases can be represented as water-on-water and the resulting contact angle will depend on the difference of the area fractions; with these assumptions the Cassie equation is reduced to  $\cos(\theta) = f1 - f2$ ,

where f1 and f2 are the fractions of two different solid phases. They show that the stability of the superhydrophobicity for the equilibrium contact angle by manufacturing a surface which remained superhydrophobic based on the difference of area fraction. Wang et al. [31] studied the static contact angle and sliding angle for systems with controlled surface temperature and humidity. They use smooth and roughened aluminum surfaces, as well as smooth and roughened PTES coated aluminum surfaces. They observed a reduction in static contact angle for all surfaces as relative humidity increased for a fixed surface temperature. The data was reported for a system with surface temperature -10°C, and increasing relative humidity from 30%, to 60% and 90%. This study brings to attention again the importance of the system conditions on the contact angles for different types of surfaces.

Based on the discussion above, two common knowledge gaps exist. The first knowledge gap is that the contact angle variation over time is not discussed in any of the literature. Condensation is a dynamic process, i.e. the condensate on the surface grows on surfaces [32-35] over time. Therefore, we could expect that the contact angle on the surface can also vary with time. The second knowledge gap is that the evaporation effect is typically ignored in the literature, which focuses on the effect of the condensation on contact angle. Evaporation is always associated with condensation [33]. Once a liquid phase condenses on the surface, it starts to evaporate. The evaporation can affect the partial pressure around the drop; hence, it can affect the condensation process. Therefore, the roles of evaporation need to be discussed.

In this work, we investigate the effect of condensation on the surface contact angle, in order to fill the two knowledge gaps discussed above. A newly developed ellipse fitting contact angle measurement method that can measure the drop contact angle without a clear view of the baseline was used to measure the contact angle of the drop [36]. A theoretical model which considered both condensation and evaporation was provided to support the discussion for the experimental observations. At the end of this paper, we demonstrate the importance of understanding the condensation effect in practice using the drop shedding by airflow as an example.

### 2. Methodologies and Materials

#### **2.1 Surface Preparation**

Three different surfaces (polystyrene, Teflon AF, and superhydrophobic) were used throughout this study. All surfaces were prepared by spin coating using a solution on an aluminum sheet (25.4 x 76.2 x 1.6 mm). Before coating, substrates were cleaned by sonication in acetone, rinsed with deionized water, and then exposed to a dry air stream. The polystyrene surface (PS) was fabricated by spin coating a 1% (w/w) solution of polystyrene in toluene (both from Sigma Aldrich) onto the substrate. The second surface (Teflon AF surface) was also prepared by spin coating a 20% (v/v) solution of tetrafluoroethylene (Dupont) in FC-40 (Acros Organics). Finally, the superhydrophobic surface, denoted SS, was prepared on aluminum substrates using in-house formulations (details can be found in [24]).

#### 2.2 Contact angle measurement

A closed-loop wind tunnel (see Figure 1) was designed, which enabled us to vary the system parameters for the study of condensation independently. Surfaces can be mounted into the test section of the tunnel during each experiment. The interface temperature, the relative humidity, and the air velocity can be controlled independently. The interface temperature can be controlled between 0°C and 70°C with an accuracy of 0.1°C by a Peltier cell driven by a PID controller with closed-loop feedback. Relative humidity of the air supply can be controlled between 10 % and 80% by mixing a flow of dry air with a fine water spray generated by an ultrasonic atomizer; a closed-loop feedback system controls the introduction of water vapor or dry air at separate inlets. Air velocity was controlled using a variable speed fan and monitored in the range between 0 and 7 m/s by a hot-wire anemometer.



Figure 1. Experimental setup for the tests.

The test section has a square inlet 55×50 mm and a length of 120 mm; a honeycomb screen with holes of 4 mm hydraulic diameter ensures uniform flow (see the insert in Figure 1). A motorized syringe is fixed to its top panel of test section to control the drop volume and perform quasi-static contact angle measurements on the test surface. The test surface assembly, including the thermoelectric cooler and a copper block, is fixed to the bottom wall of the test section; four thermocouples are inserted into the copper block (see Fig. 1) to measure the temperature distribution.

The ambient temperature in the tunnel was fixed by lab conditions at an average of 25 °C. The air velocity was kept in the range of 1 m/s to help homogenize the vapor distribution in the tunnel and to replenish the flux of vapor that condensates at the test section. The contact angle measurement was conducted under both high and low relative humidity conditions. The high humidity condition

measurement represents the condensation environment. To achieve condensation on the test surface, the surface temperature was decreased to 10°C with the relative humidity of 45% (inside the wind tunnel). The contact angle measurement was conducted after the surface was exposed for 5, 22, and 45 mins (representing different condensation times), respectively. For each measurement, a water drop was generated on the surface using a syringe. The drop was then expanded by adding water to the drop for measuring the advancing contact angle and contracted by withdrawing water for the receding contact angle measurement. After each of the tests, the surface was heated to dry.

An ellipse fitting method that fits the drop profile with a general equation of an ellipse whose major and minor axes are not necessarily parallel and normal with the substrate, respectively, was used for contact angle analysis. This method is shown to be able to use the visible part of the drop profile to estimate the contact angle of a drop without a clear view of the baseline (for detail of the ellipse fitting method see [36]). Between heating and condensation stages, the surface was not moved, preserving the location of the baseline on the surface. For each surface, contact angle measurement was also conducted under the low relative humidity condition (20%) for reference. The low humidity measurement serves two purposes. Firstly, it generates the benchmark data for understanding the condensation effect on surface contact angle. Secondly, for the high humidity measurement, the condensed drop in the foreground and background will block the view to observe the details around the drop baseline during the contact angle measurement. Therefore, the information obtained through the low humidity measurement could help to understand the observation in the high humidity measurement.

#### **2.3 Drop shedding test**

The drop shedding test was conducted using the wind tunnel described in the previous section. For the drop shedding test, a surface with known wettability was mounted into the tunnel's test section for each of the measurements. An overhead camera (Prosilica GT1600, Allied Vision) was used to take images of the top view. For each test, the wind tunnel is first set to be in the wet condition (45% humidity and 1.3m/s) to allow condensation. The air velocity was kept in the range of 1 m/s to help homogenize the vapor distribution in the tunnel. Once the condensed drop reached our target volume, airflow speed was increased until the motion of drop on the surface can be seen.

We record the rate of the airflow when the drop starts to move. The tunnel airflow is characterized as shown in [37]. The air dynamic within the wind tunnel is beyond the scope of this work, therefore it is not discussed here.

## 3. Results and discussion

#### 3.1 Changes of contact angle over time

Figure 2 displays the advancing and receding contact angle values measured on the Polystyrene, Teflon AF, and SS surfaces as a function of time. A general decreasing trend of contact angle over time (both advancing and receding contact angles) can be seen for all the three surfaces. The most significant change of contact angle over time occurs on the SS surface. After placing the SS surface under the condensation environment for 45 mins, the advancing and receding contact angles decreased 18.5° and 28.2° with respect to the low humidity measurement, respectively.



Figure 2. Advancing and receding contact angles for (a) Polystyrene, (b) Teflon AF (c) and SS surfaces for both low and high humidity measurement. No condensation represents the measurement results for low humidity measurement. The 5-, 22- and 45-minute measurement results represent the measurement for high humidity measurement.



Figure 3. Ring effect for water on a Teflon surface with condensation local to the droplet observed.  $\theta_{avg} = 108.2$ , surface temperature of 10 °C, relative humidity of 20%, ambient temperature of 25.8 °C.

The trends displayed in Figure 2 should be the result of condensation, snapshots of the drop during the contact angle measurement were checked. Interesting observation was made in the low humidity environment test. The snapshot of drops on Teflon AF surface under low humidity measurement are also shown in Figure 3a (t=0.5s - t=15s). A clear ring of condensation area (shadow area around the drop) can be observed during the measurement (e.g., see the snapshot at t=9s). This condensation ring becomes wider over time (see t=11s). This ring condensation on the surface can still be seen even after the drop was removed, e.g., t=15s. Such condensation ring was observed for all surfaces during the low humidity measurement.

Condensation only occurs around the drop, suggesting that the vapor close to the drop becomes saturated. Since the surface temperature is constant during the measurement, the water vapor pressure around the drop should be higher than the other part of the measurement chamber. As indicated in literature, the increase of the water vapor pressure can increase the adsorption of the water by solid surface, therefore, cause a decrease of the contact angle. To further confirm the fact that the vapor close to the drop can become saturated, a pendant drop test was performed (shown in Figure 3b). A water pendant drop, which is approximately 2mm above the cold surface, was generated under the dry environment. A condensation film appeared immediately after the drop had been generated. Similar to Figure 3a, the condensation on Figure 3b also grew over time. The new condensed film (drops) started to disappear (evaporation) when the main drop was removed

(start from t = 150s). Since there is no direct contact between the water drop and the solid surface, the condensation can only be caused by the increase of the water vapor pressure locally which is caused by the evaporation of the drop.

The formation of the condensation ring during contact angle measurement and pendant drop tests suggests that the evaporation from the drop condenses on the surface; the size of the diffusion layer for the droplet relates to the condensation ring size and existence. The partial pressure of vapor in the diffusion layer is greater than the equilibrium vapor pressure about the drop and even be greater than the dew vapor pressure at 10°C (the surface temperature). This is the reason causing the ring condensation occurs in the low humidity environment.

Therefore, it is intuitive to ask if the decrease of the contact angle over time under high humidity condition (observation made in Figure 2) was also caused by the vapor pressure to increase around the drop. That is during the condensation measurement, the evaporation from the main drop as well as the new generated condensed water drop can evaporate and cause the vapor pressure increase and lead to a decrease in contact angle. In the next Section, we try to use a modelling approach to investigate this hypothesis.

#### 3.2 Theoretical model for partial vapor pressure changes in condensation condition

In the previous section, we have shown that for the low humidity measurement, the evaporation from the main drop can cause an increased vapor pressure around the drop; hence causing the decrease of contact angle. It is difficult to provide direct evidence to prove our hypothesis, the theoretical model developed here is to show how the evaporation of the main drop can affect the partial pressure of water vapor. The system considered is shown in Figure 4: a sessile drop surrounded by a uniform (for simplicity) diffusion layer of thickness d. The density of vapor in the diffusion layer is greater than the equilibrium density of vapor for a given ambient temperature (T) and pressure (P) due to the evaporated flux (J). The diffusion layer (assumed constant in time and uniform) has a larger partial pressure than the equilibrium vapor pressure away from the droplet.



Figure 4. Schematic of a sessile drop on a cooled surface with vapor region around the droplet (due to evaporation of the droplet); the vapor density is a function of distance from the droplet. The integration of the vapor density (curve blue) is shown by the box at a certain distance for the diffusion layer. Condensation local ring for a sessile droplet on a cooled surface is also shown in the schematic.

To develop the mathematics for our hypothesis, we assume that the evaporated mass stays within the diffusion layer with a constant thickness; we also assume the diffusion layer is uniformly dense. Next, we integrate the change in vapor density at the surface of the droplet to the equilibrium vapor density of the environment. The evaporation and condensation phenomena were decoupled; calculations do not involve re-condensation of the evaporated mass. To calculate the flux, the work of Chini and Amirfazli [38] was used, i.e. Eq.1 and Eq.2. To calculate the pressure within the diffusion layer and the thickness of the diffusion layer, Eq.3 and Eq.4 were derived. Equation 1 is arrived at by adaptation of a Maxwellian model (assumes the evaporation process is quasi-steady) for the evaporation of a sphere suspended in quiescent environment i.e., using steady-state and non-convective assumptions. This model as a first approximation is extended to sessile drops with the assumption of it being a spherical cap; this is valid for micro-liter droplets with a low bondnumber. The expression in Eq.1 describes the effect of the solid substrate on the evaporation rate is accounted for by a contact angle correction parameter, Eq.2, which is a closed form fitting equation derived in Ref. 38. Functionally, the contact angle correction parameter accounts for the change in total evaporation due to the substrate. Physically it represents that the evaporation along the free surface of the sessile drop is non-uniform unlike the suspended sphere.

The equations below describe the relationship between the pressure in the diffusion layer and the evaporated flux of the droplet  $J_D$  (the detailed derivations of the equations are provided in the Supplementary Information).

$$J_D = \frac{-2\pi \cdot R \cdot \rho_{vs} \cdot D}{s} \cdot \left( w_{eq} - w_{\infty} \right) \cdot \frac{\sin(\theta)}{1 + \cos(\theta)} \cdot f(\theta)$$
(1)

$$f(\theta) = 56.561 \cdot \theta^{-0.892} \tag{2}$$

$$P_{\nu,\delta} = P \cdot \frac{\left[\frac{P_{\nu0}}{P - P_{\nu0}} + \frac{J_D \cdot R \cdot T_d \cdot t}{18\delta \cdot (P - P_{\nu0})}\right]}{1 + \left[\frac{P_{\nu0}}{P - P_{\nu0}} + \frac{J_D \cdot R \cdot T_d \cdot t}{18\delta \cdot (P - P_{\nu0})}\right]}$$
(3)

$$\delta = D \cdot \frac{\rho_{tot}}{J_D} \cdot \ln\left(\frac{\rho_{tot} - \rho_{vs}}{\rho_{tot} - \rho_{vo}}\right) \tag{4}$$

The evaporated mass was allowed to accumulate in a constant and uniform diffusion layer without condensing on the surface (This will represent an upper bound for vapor pressure in the diffusion layer). If the pressure increase in the diffusion layer of a droplet in this scenario is high enough, then condensation and film formation (ring condensation as shown in Figure 3) local to the droplet and independent of the equilibrium vapor pressure of the environment can be seen.

Eq.3 quantifies the effect of the partial vapor pressure of the water is used to leading to formation of a condensation ring. The average volume of the drop on the surface under the high humidity condition for 5mins is 2.5nl, 1nl, and 0.7nl for PS, Teflon AF, and SS surface, respectively.

Figure 5 shows how much vapor can accumulate (due to evaporation) in the diffusion layer by a 2.5nl (on PS), 1nl (on Teflon AF), and 0.7nl (on SS) drop, respectively. A clear increase of the vapor pressure over time can be seen for all the surfaces. Based on Figure 5, one can know that the evaporation phenomena in time contributes to the vapor pressure increase in a shell of vapor surrounding the droplet. The increase in pressure around a droplet can further cause a condensation ring around droplets on cooled surfaces.



Figure 5. Vapor pressure around a single droplet as a function of time, vapor is allowed to accumulate in a constant and uniform diffusion layer. The volume (for PS, Teflon AF and SS is 2.5nl, 1nl and 0.7nl) and contact angle of drop, for calculation are taken as the average at 5 minutes of condensation with an interface temperature of 10 °C. Relative humidity is 45%, ambient temperature is 25.8 °C.

Using the above modeling approach, a parametric calculation was carried out. Figure 6a presents the vapor pressure in the diffusion layer for 50 droplets with different volumes one second after deposition. It can be seen that for drops smaller than 1.2  $\mu$ L, pressure is larger than the dew point pressure, suggesting a local condensation can occur due to the main drop. The partial pressure for all drops increases over time. When the drop was placed on the surface after 5s (Figure 6b), even the largest drop (5  $\mu$ l) has a partial pressure larger than the dew point pressure. This suggests that even in the high humidity environment, the condensation ring can be formed. It should be pointed that this model is a first-order approximate model, to demonstrate that it is theoretically possible for the condensation occur on the surface caused by the drop evaporation. Several made assumptions make this a first order model, e.g., the diffusion layer is homogeneous, the evaporated water vapor will condense on the surface. However, the model can explain the reason for the

observation in Figure 2. During the condensation process, the number of condensed drop increases, hence, provides a thicker diffusion layer (close to the solid surface) with a higher vapor pressure. The partial pressure of water vapor will then increase the adsorption. Since all the three surfaces used in this work PS, Teflon AF and SS have relatively low surface free energy, the adsorption of water can increase their surface free energy at the interface; hence causing a decrease in the contact angle [30,31]. For the SS surface, the absorption can cause the surface to lose its super-hydrophobicity; hence the most significant change of the contact angle among the three surfaces was seen in Figure 2.



Figure 6. Results of the developed model for vapor pressure due to evaporation of different volume water droplets of different volume on a Teflon surface with  $\theta = 108.2$  after 1 second (a) and 5 second (b) of evaporation. The surface temperature is 10 °*C*, relative humidity is 20%, and ambient temperature of 25.8 °*C*.

#### 3.3 Drop shedding by airflow

So far, we have understood how the contact angle varies during the condensation process. In this section, we use the drop shedding problem to demonstrate the importance of understanding the condensation effect on the drop contact angle. Figure 7 shows a typical shedding process of water from a solid surface (Teflon AF). The Teflon AF surface was first exposed under the high humidity condition for 45mins to allow the drops to condense on the surface. The air velocity was then increased to shed the drops away from surface. The first drop on the upstream [39, 40] started to move when the air velocity increased to approximately 5.4 m/s (critical velocity). When the drop moves, it coalesces with the small drops on its shedding path. As it coalesces, the volume increases during the shedding process.





The critical velocities for all the three types of surfaces are summarized in Figure 8 after being exposed under high humidity environment for 5mins, 22mins and 45mins, respectively. The values of the drop volume shown in Figure 8 is the volume of the first shedding drop for each test. It can be seen that for the drops condensed on one surface, the critical velocity increases with the increase of condensation time. This is because the volume of the condensed drop increases over time. For example, the volume of the condensed drop on SS surface increased from 0.8nl to 55nl when the condensation time increases from 5 mins to 45mins. The increase of the volume leads to a decrease of the critical shedding velocity similar to what was reported in [24] for icing conditions.

More interestingly, for the cases with the same condensation time, the largest  $U_c$  among the three surfaces is different when the condensation time changes. For the 5 mins condensation case, the Uc for the SS surface is 12.1m/s which is smaller than that of the Teflon AF and PS surfaces. However, when the condensation time increases to 45 mins, the critical velocity for SS (8.5 m/s)becomes larger than that of Teflon AF (5.8m/s) and PS (5.4m/s) surfaces. This is because of the change of CAH (the difference between advancing and receding contact angle) over condensation time (see Figure. 2). Besides the drop volume, the CAH is another important parameter governing the critical velocity. As discussed in Ref. 24, the value of Uc increases with the increase of surface CAH. The value of CAH for SS increases from 9.3° to 18.2° when the condensation time increases from 5mins to 45 mins (see Figure 2). However, for the Teflon AF and PS surface, the CAH value is either decreasing or flat with the increase of condensation time. The CAH (18.2°) for SS surface at 45mins condensation is the largest among the three surfaces. The example for drop shedding shows the importance of understanding the condensation effect on the contact angle over time. Under the dry condition, the SS requires the smallest  $U_c$  to blow away the drops from the surface. However, under the condensation condition, since the CAH of SS increases significantly, it requires the largest  $U_c$  value for shedding.



Figure 8. The critical velocity  $(U_c)$  for shedding the condensed drops from solid surface after 5mins, 22mins, and 45mins condensation, respectively.

### 4. Conclusion

The effect of condensing on the contact angle value of sessile drops was studied in this work. A general decrease of contact angle over condensation time was found for all three surfaces examined. A condensation ring around the drop on the surface was observed. This suggests the vapor pressure around the drop is larger compared to surrounding. It was found that the change of contact angle over time is caused by the evaporation of the drop which forms the condensation ring and an adsorbed.

The experimental observations were explained through a newly developed first order model. To our knowledge, this is the first model which considers the roles of evaporation for the study of condensation effect on drop contact angle. The model results showed that for the high humidity environment, the drop evaporation can cause the increase of the vapor pressure locally. The increase of the vapor pressure can further lead to the formation of a condensation ring around the drop causing a decrease in the contact angle in time. We also demonstrated the importance of understanding the condensation effect on contact angle using the problem of drop shedding under the condensation condition as an example. It was shown that after being exposed to the high humidity condition for 45 mins, the SS surface requires the largest Uc for shedding away the condensed drop, compared with the PS and Teflon AF surfaces. This is because of the significant increase of CAH value for SS surface under the condensation environment.

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