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ON THE INITIAL DROPLETS FORMATION DURING THE PROCESS OF DROPWISE CONDENSATION (I)

滴状凝縮過程における初期液滴の発生について by Ichiro TANASAWA* and Shin-ichi NAGATA*

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1. Introduction

Observing the surface on which dropwise condensation is taking place, one can see the each individual drop undergoing successive life-cycles composed of four subprocesses; formation of the initial droplets, growth by condensation, growth by coalescence, and detachment from the surface (Fig. 1).

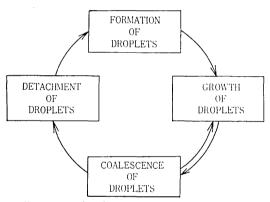


Fig. 1 Cycle of the Dropwise Condensation.

As the first step of the systematic research program on dropwise condensation aiming to fully understand the secret of heat-transfer mechanism and, eventually, to find the key to the practical application of this very efficient process to actual heat transferring devices, the present authors and his colleagues have developed the method of simulating the entire process of dropwise condensation using digital computer. 1)2) They also have published the results of application of the above mentioned method to the process of drop growth due to coalescences with neighboring drops.3)4) Further, they have been conducting experiments on dropwise condensation intending to obtain data necessary for the simulation.5)

In the present report the attention is focussed on the process of the initial droplets formation during dropwise condensation, which is to constitute a different side of the research program.

Concerning the machanism of the initial droplets formation, Jakob⁶⁾ first proposed the "film-fracture theory". He considered that the vapor molecules condense onto the bare surface between drops forming a thin film of liquid. This film grows until it reaches a critical thickness when it "rolls itself together and fractures" to form droplets. Some thirty years had ellapsed when Welch and Westwater73 made high-speed micro-cinematographic observation of the condensing surface, and found the fact that, whenever two or more drops coalesced, the part of the metal surface previously covered by the drops was exposed and a distinct flash of light from the lustrous bare metal was observed, and that the lustre was seen to fade after a few milli-seconds. They estimated that the disappearance of the luster indicated the presence of the liquid film. They calculated the critical film thickness from the time necessary for the (seeming) film formation and the amount of heat transferred meantime and obtained $\delta_c = 0.13$ $\sim 1 \,\mu$. Ruckenstein and Metiu⁸⁾ obtained $\delta_c = 0.27$ μ as a consequence of a thermodynamical consideration supplemented by the experimental result of Welch et al. In Japan, Sugawara and Katsuta9) observed the process of dropwise condensation of humid air onto a metalic surface through interference microscope and measued the distance between interference stripes just before the initial drop appearance, obtaining $\delta_c = 0.63 \,\mu$.

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Contrary to the film-fracture hypothesis as demonstrated above. Tamman and Boehme¹⁰⁾ had suggested that the droplets always originate from the fixed nucleation sites randomly distributed over the surface. This "nucleation hypothesis", however, had not been adopted for a long time because of the experimental observation by Welch et al, or the theoretical analysis by Ruckenstein et al. A few years ago, Umur and Griffith¹¹⁾, based on thermodynamic considerations together with a skillful optical measurement utilizing the polarized light technique, supplied a piece of evidence which strongly proves that a film no greater than a monolayer thick can exist in the area be-This work proved that droplet tween drops. formation is a result of a nucleation process. Thenceforth Peterson and Westwater¹³⁾ and Mc-Cormick and Westwater¹²⁾ have supported the nucleation hypothesis in their works.

The present authors also approve the nucleation hypothesis, as already have demonstrated in their papers. Although the rightness of the nucleation hypothesis seems to have no slightest shadow of doubt, there are still a few who are inclined to to support the film-fracture hypothesis. It might be chiefly because the initial droplets are so minute as to be observed by means of the ordinary optical observation technique and the inconteslable proof is hard to obtain.

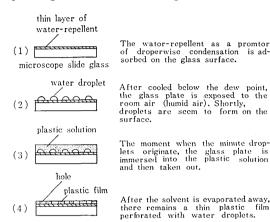
The authors happened to know that the initial droplets originated on a solid surface could be observed, though indirectly, utilizing an electron microscope technique. They thought that it should be possible to throw light on the process of the initial drop nucleation and to offer some definite evidence for the nucleation hypothesis. The present report is a summary of the preliminary results.

2. Method of Observation

The procedure described in the following is devised by Fukami and Adachi¹⁴⁾ as a new method of preparating micro plastic nets useful for correction of astigmatism of an electron microscope

of high magnification. Their object was, however, to prepare micro plastic nets of small holes for specimen supporting, while the present authors intended to utilize the technique for observation of the sizes and the distribution of the initial droplets formed on the condensing surface.

The outline of the method of preparing a micro plastic grid is illustrated in Fig. 2.



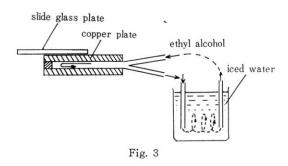
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The perforated plastic film is separated from the glass surface with the aid of a re-hydrophilic agent.

Fig. 2

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If we describe in more detail the each stage of the method in Fig. 2: (1) A microscope slide glass of excellent quality (about 0.8 mm in thickness) is used as a condensing plate. At first, the glass plate is completely cleaned with high purity carbon-tetrachrolide, and dried. Then it is immersed into the 0.03% aqueous solution of the water-repellent, Softex KWO (distearyl dimethyl ammonium chloride, Kao-Atlas Co.) for about 30 minutes or more, and is taken out, washed with distilled water to remove excess water-repellent, and is dried. (2) This slide glass plate must be cooled to the optimum temperature for the formation of droplets. The precedure employed by the authors is shown in Fig. 3. Ethyl alcohol cooled to the previously determined temperature by flowing through iced water is induced between the gap of a thin box composed of two copper plates and spacers, keeping the surface temperature

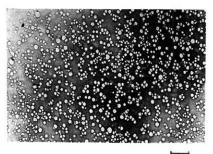


of one of the copper plates lower than the dew point by about 0.5 degC. After dews attached to the copper plate are wiped off, the glass plate prepared in (1) is put on for a very short period. (If this period is too long, the droplets formed on the glass surface grow too large, while, if too short, no droplets are observed to form. In the present experiment, the period of three seconds was found to be most adequate.) (3) The glass plate carrying newly formed droplets is quickly immersed into Triafol solution (0.1% solution of cellurose acetobutyrate in extra pure ethyl acetate), then taken out and tilted to let the excess solution flow away. (4) While the solvent, ethyl acetate, is rapidly evaporating, secondary dews often originate because the latent heat for evaporation is taken off. To prevent this phenomenon (called brushing) an infrared lamp is used to heat the surface slightly. (5) In order to remove the perforated plastic film from the glass surface in good condition, the plate is immersed into the rehydrophilic agent, 0.5% aquaous solution of Pelex OTP (sodium dialkyl sulphosuccinate, Kao-Atlas Co.) for a few minutes. Then it is taken out and washed with water. The glass plate kept almost horizontally is slowly put into the water and then Triafol micro plastic film is separated, comes floating onto the water surface. (6) The micro plastic grid floating on the water surface is scooped up with the metal grid for electron microscope use, fixed on it and dried. As the Triafol grid thus obtained is rather weak, it is recommended to reinforce it by evaporating carbon in vacuum onto its surface.

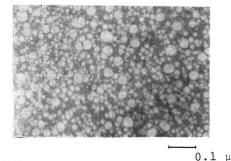
3. Results of Observation

In case when the micro grid is used for its own original purpose, it is requested to prepare the plastic film having small holes of previously determined radii, while, in the present experiment, the object is to observe the minimum radius and the distribution of the droplets formed on the glass plate under a known temperature condition.

In Fig. 4, examples of pictures taken through an electron microscope are shown. In case of Fig. 4(a), the glass plate was exposed to the humid air for rather long period (about 7 seconds) and the initial droplets grew to $0.1\sim0.2\,\mu$ in radii.



(a) magification: 5600 1 μ dew point: 15 °C exposure time: 7 s



(b) magnification: 92,000 dew point: 10 °C exposure time: 3 s

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In Fig. 4(b), the exposure time was about 3 seconds
which was approximately the minimum period
below which no initial droplets could be observed.
The diameters of the smallest holes seen in the
picture are around 100 A, and the number density
of the distinguishable holes is counted as about

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2600 per square micron (or 2.6×10^{10} /cm²). In both cases, the difference between the saturation temperature of the water vapor (the dew point) and the surface temperature of the glass plate was, at its maximum, 0.5 degC. A thermodynamical calculation assuming the equilibrium state yields about 680 A as the critical diameter of the droplet which can grow on the plate under the above mentioned temperature difference. Nevertheless, none of the holes seen in Fig. 4(b) exeed the critical value (the largest one being nearly 400 A in diameter). At present it cannot be determined definitely whether the droplets actually having diameters less than the critical value are taken in the picture or the micro plastic films may have a characteristic that the holes perforated through them exhibit smaller diameter than that of the droplets formed on the glass plate.

To proceed on with further quantitative discussion, a few improvements will be neccessary such as on the more precise measurement and control of the surface temperature. However, it may at least be concluded so far that the origination of droplets on the glass plate is due to nucleation process, and that the population density of nucleation site is, at the lowest, of the order of 10¹⁰ per square centimeter.

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