



# Dropwise condensation rate of water breath figures on polymer surfaces having similar surface free energies

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## ABSTRACT

This study investigates the effect of surface roughness, wettability, water contact angle hysteresis (*CAH*) and wetting hysteresis (*WH*) of polymeric substrates to the water drop condensation rate. We used five polyolefin coatings whose surface free energies were in a close range of 30–37 mJ/m<sup>2</sup> but having different surface roughness and *CAH*. The formation of water breath figures was monitored at a temperature just below the dew point. The initial number of the condensed droplets per unit area ( $N_0$ ) and droplet surface coverage were determined during the early stage of drop condensation where the droplet coalescence was negligible. It was found that the mean drop diameter of condensed droplets on these polymer surfaces grow according to a power law with exponent 1/3 of time, similar to the previous reports given in the literature. It was determined that surface roughness and corresponding *CAH* and *WH* properties of polymers have important effects on the number of nucleation sites and growth rate of the condensed water droplets.  $N_0$  values and the surface coverage increased with the increase in surface roughness, *CAH* and *WH* of the polymer surfaces. The total condensed water drop volume also increased with the increase in surface roughness in accordance with the increase of the number of nucleated droplets.

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## 1. Introduction

If a condensate does not completely wet a surface, dropwise condensation occurs and many droplets whose diameters ranging from a few nanometers to micrometers occupy the condensing area. Dropwise condensation is desired in most of the heat transfer applications because thermal conductivity greatly increases due to the absence of a continuous film on the condensing surface [1–4]. When we breathe on a hydrophobic surface, a fog of tiny droplets named as “breath figures” forms [5–16]. The number of condensed droplets per unit area and mean droplet sizes vary according to the properties of the solid surface. This indicates the important role of substrate surface free energy, wettability and contact angle hysteresis on the behavior of condensed water droplets [8–11,16–22]. Most of the condenser surfaces are generally made of metals in heat exchangers, and metals usually exhibit filmwise condensation rather than dropwise because of their high surface free energies and require surface modification to shift into dropwise condensation [23–25]. Polymeric or organic coatings were used on metals as “promoters” to reduce their surface free energy and to obtain dropwise condensation in industry [23,26–28].

In general, dropwise condensation consists of a combination of several processes: numerous minute droplets are formed after the vapor impinges on a surface cooled to a temperature below the saturation temperature, releasing the latent heat of condensation and these droplets grow very rapidly due to the continuing direct condensation of vapor onto them. Some of the droplets touch each other and coalesce to form larger drops and droplets shift their positions a little at each coalescence, leaving open areas on the surface where initial droplets can be nucleated again. It is almost impossible and does not make sense to attempt to treat the nucleation and coalescence steps separately [6–13,16]. In most of the published drop condensation studies, flowing water vapor at a specific humidity was directed onto cool surfaces causing rapid condensation [6–10,12,14–16,29,30]. Nevertheless, non flowing vapor experiments, i.e. condensation of water vapor from ambient air was also studied [11,13,21,22]. The early stage of condensation where the droplet coalescence is negligible was the less frequently studied stage of droplet growth [8,9].

The “surface coverage” parameter was used in most of the droplet condensation studies and defined as the ratio of surface area covered by the droplets over the total substrate area in order to quantify the droplet growth by optical microscopy photographs. This quantity varies while drop perimeter grows and other droplets nucleate by time depending on the growth, coalescence and re-nucleation steps [6–16,31]. Surface coverage change by dropwise condensation was investigated on silanized glass [6,7,16],

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various polymer [8,9,11,22], and patterned superhydrophobic model surfaces [12,13,29,30,32–37]. It was reported that the magnitude of surface coverage and the extent of dropwise condensation are dependent on the surface properties of the substrate such as surface tension [19,20,26–28,31], initial contact angle [10,11,16,22], and contact angle hysteresis (CAH) [10,16,17].

Contact angle is the angle formed by a liquid drop at the intersection point of three-phase boundary and it is included between the tangent plane to the surface of the liquid and the tangent plane to the surface of the solid. Equilibrium contact angle,  $\theta_e$ , is a quantitative measure of the wetting of a solid by a liquid and it is a manifestation of the thermodynamic equilibrium of the three-phase system [38]. Low values of contact angles indicate a strong liquid–solid interaction and the liquid tends to spread on the solid surface. The increase in surface coverage with the decrease in contact angle and corresponding heat transfer efficiency was reported in some of the articles for a broad range of contact angles [10,11,16,28].

If an ideal surface (chemically homogeneous, rigid, and atomically flat) is present, there would be a unique equilibrium contact angle  $\theta_e$ . However, as a consequence of the non-ideal nature of any real surface, the presence of surface roughness, chemical heterogeneity and surface restructuring can cause to measure different contact angle values on a surface. When a liquid drop is formed on a substrate surface by injecting the liquid from a needle connected to a syringe, it is allowed to advance on the fresh solid surface and the measured angle is called as “*advancing contact angle*”,  $\theta_a$ . The other contact angle type is the “*receding contact angle*”,  $\theta_r$  and it can be measured when a previously formed sessile drop on the substrate surface is contracted by applying a suction of the drop liquid through a needle. Contact angle hysteresis, CAH on a surface is the difference between advancing,  $\theta_a$  and receding,  $\theta_r$  contact angles ( $CAH = \theta_a - \theta_r$ ) and is a measure of the surface roughness and chemical heterogeneity of the surface [38–40]. The wetting hysteresis (WH) is defined as the adhesion energy to cause contact angle hysteresis [ $WH = \gamma_{LV}(\cos \theta_r - \cos \theta_a)$ ]. CAH was regarded as an important factor to determine the efficiency of the drop condensation process [2,6,10,16,17,19,20,29]. In these reports, authors generally investigated CAH effect on the droplet coalescence [2,6,10,29] and on heat transfer efficiency of a surface and found that a decrease of CAH results in high heat transfer at the last stage of condensation because large droplets easily flows on surfaces having low CAH [16,17,19,20]. CAH effect on the growth of the condensation pattern was reported by Zhao and Beysens and it was found that the contact-line-pinning on the chemically heterogeneous surface prevented the full coalescence of droplets, and the saturated surface coverage is significantly increased depending on the hysteresis strength [10]. Beysens and coworkers showed that ellipsoidal and not hemispherical droplets form after coalescence by the CAH effect and the surface left free by coalescence may not be large enough to compensate for the growth and preserve scaling and thus surface coverage increases continuously; interconnected droplet structures form, leading to pseudo-wetting films [2,6,29]. Neumann and coworkers presented detailed experimental results on CAH effects on drop condensation and concluded that heat transfer strongly depends on CAH during the dropwise condensation and increases with decreasing CAH [17]. The decrease on the mobility of the condensed droplets and the efficiency on the condensation heat transfer with the increase in CAH were reported by Ma and coworkers [19,20]. However, none of these articles investigated CAH effect on the formation of breath figures in the early stage of drop condensation. On the other hand, the variation of CAH on patterned model surfaces depends on the size and separation distance between the pillars [32–37,41,42]. Wier and McCarty showed that water droplets nucleated and grew both on top of and between the pillars on ultrahydrophobic surfaces

and when condensation progressed, water between the pillars was forced upward to the surface. Cooling this sample below the dew point resulted in a decrease in  $\theta_r$  down to  $0^\circ$  and an increase in CAH. Condensed droplets pinned at the contact lines and water drop mobility reduced on the patterned surfaces [32].

In this study, we monitored the formation of the water breath figures for the early stage of drop condensation where the droplet coalescence is negligible in ambient conditions on horizontal polyolefin surfaces whose surface temperatures were just below the dew point. Our aim is to investigate the effect of surface roughness and corresponding CAH and WH on the surface coverage and the rate of slow dropwise condensation of water vapor from ambient air when surface free energies of the surfaces are in a close range. We especially preferred to study the drop condensation in this stage in order to achieve better discrimination of the surface roughness effects. We used five different polyolefin surfaces having similar surface free energies in a range of 30–37 mJ/m<sup>2</sup> having different surface roughness. Initially, it was found that the mean drop diameter of condensed droplets on these polymer surfaces grow according a power law with exponent 1/3 of time as reported previously in the literature where the droplet coalescence is negligible. It was also determined that surface roughness, initial contact angle, CAH and WH effects are important on both the nucleation and drop condensation rates. The results are discussed throughout the text.

## 2. Experimental

### 2.1. Materials

Commercial polypropylene (PP) was purchased from PETKIM, Turkey (PETKIM MH 418); ethylene–vinyl acetate copolymer containing 12% vinyl acetate content by weight (EVA) was provided by Dupont (ELVAX 660); high density polyethylene (HDPE) was provided from Basell Inc. (HOSTALEN-GM8255), polypropylene–polyethylene copolymer elastomer containing 12% polyethylene content by weight (PPPE) was purchased from Dow Chemical Co. (VERSIFY 2300) and low density polyethylene (LDPE) was supplied from PETKIM, Turkey (F5-21T). These polymers were used as received to prepare polymer solutions in technical grade xylene solvent (TEKKIM, Turkey). 76 mm × 26 mm standard glass slides (ISOLAB, Turkey) were coated with the above polymers by dip coating technique [43]. Two-component adhesive polyepoxide layer (404 Chemicals, Turkey) was applied as the primer coating onto glass slides to compensate weak adherence of polyolefins. Spectroscopic grade water, methylene iodide,  $\alpha$ -bromo naphthalene, ethylene glycol and formamide liquids (all purchased from Merck) were used to form droplets where contact angles were measured to calculate solid surface free energy.

### 2.2. Sample preparation

76 mm × 26 mm standard glass slides were initially cleaned in chromic acid, rinsed with distilled and Milli-Q<sup>®</sup> water, and dried in a vacuum oven. PP, HDPE, PPPE, LDPE and EVA polymers were dissolved in xylene between 60 and 130 °C to obtain solutions from 20 to 40 mg/ml (w/v). Polyepoxide coated glass slides were prepared from its chloroform solution and were dip-coated in these polymer solutions at specific temperatures and dipping rates by using a mechanical dipper. Specific dipping solution temperatures were 102, 115, 100, 130 and 105 °C for PP, HDPE, PPPE, LDPE and EVA samples. Specific glass slide dipping rates were 77, 57, 360, 320 and 612 mm/min for PP, HDPE, PPPE, LDPE and EVA samples. Chemically heterogeneous surfaces having varying surface roughness were formed from copolymers by phase segregation during this controlled dip coating and solvent evaporation process [43–46].

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