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Effects of insoluble nano-particles on nanofluid droplet evaporation

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ABSTRACT

A model is presented for predicting the evaporation behavior of liquid droplets containing nano-sized insoluble particles. The model is constructed based on, and supported by, the existing experimental result. Of particular interest are the evolution of droplet size, and evaporation rate constant prior to the shell formation (also called the first drying or evaporation stage), when the particles forms a compact shell at the droplet surface. As the droplet evaporation proceeds, increasingly more particles are "included" on its surface, reducing the effective liquid surface area for evaporation causing deviation from the classical d^2-law for pure fluid droplet evaporation. Peclet number (Pe), a non-dimensional ratio of particle diffusion time to the droplet lifetime, and initial particle concentration (Y_{vo}) are shown to play a role in the evaporation behavior prior to shell formation. The model predicts that: (1) the deviation from the classical d^2-law is increasingly significant with increased Pe and Y_{vo}; (2) the time to shell formation decreases, and shell diameter increases, with increased Pe and initial particle concentration (Y_{vo}). As a consequence, $Y_{vo} \cdot Pe < 1.8$ is necessary for the first stage to exist at all. Within the limiting value, the combination of larger Pe and smaller Y_{vo} produces larger hollow shells with low densities. For $Y_{vo} \cdot Pe > 1.8$ the shell forms instantly and evaporation occurs after liquid diffuses through, and wet, the shell. The effect of contact angle (θ) on the effective liquid surface area for evaporation is also discussed.

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1. Introduction

Evaporation of liquid droplets finds a wide range of applications—pharmaceutical, food, spray drying, energy, and propulsion fuels among others. The evaporation in stagnant as well as flowing surrounding gases has been studied both experimentally and analytically [1–8]. Evaporation of pure liquid droplets in the diffusion-controlled regime is known to follow the so-called d^2-law,

$$d^2 = d_0^2 - Kt \tag{1}$$

where d and d_o are the instantaneous and initial droplet diameters, respectively, and K is the evaporation coefficient. That is, the evaporation rate is directly related to the surface area that absorbs heat through diffusion. Droplets in convective environment does not follow the d^2-law, as the temperature and flow fields cease to be spherically symmetric and the exact value of exponent becomes a function of parameters such as Reynolds, Grashof, and Nusselt numbers. In general, the droplet evaporates follows a d^n relationship [9],

with $n = 2$ for a diffusion-controlled evaporation in quiescent environments. Multicomponent droplets also exhibit evaporation behaviors according to the d^2-law that are analogous to a batch distillation [10], with varying values of K as evaporation proceeds through different stages.

Another type of multicomponent liquid droplets consist of those containing insoluble particles (see for example [11–13]). For these types of droplets, the initial (i.e., first stage) evaporation rate was found to correspond to that of pure liquid, followed by formation of the shell of particles [11]. The shell forms when the droplet surface is populated with solid particles and its size is assumed constant through the rest of the process [11–16]. The second period of evaporation commences after shell formation, where evaporation occurs when the liquid from within the shell reaches the surface of the shell through capillary action. The second stage comprises essentially of drying of the wet particle agglomerate. A conceptual characteristic drying curve (CDC) was proposed to describe the heat transfer phenomena throughout the entire drying process [14]. Some experimental evidence supports the constant shell diameter [11].

For the purpose of the present study, it is worth noting that the particle agglomerate at the completion of drying possesses a wide

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range of morphology, from shallow crust (i.e., a shell with void inside) to compact cluster. In a study of water droplets containing nano-sized alumina particles, the morphology of dried particle agglomerate depends on the initial volume fraction of the colloids in the droplet and the mobility of particles within the liquid [17]. Derkachov et al. [18] in a study of evaporation-driven aggregation of 200-nm particles found that the morphology of nanoparticle aggregates ranges from spherical shells to the solid-like microcrystals. Such a wide range of morphological variations is attributable to effects including evaporation rate for a given initial concentration investigate (1:1000 polystyrene spheres in water). The detail morphology of the final particle aggregates involved detail study of contact mechanics [19] and is beyond the scope of this study.

To explain the vast morphological variations, Derkachov et al. performed a numerical simulation [18] to investigate the *inclusion* process (the capturing of nanoparticles on the regressing droplet surface). The results reveal that the particle density on the regressing surface increases with time while forming "islands". As a consequence, the surface area from which evaporation occurs might be smaller than that determined by the droplet geometry depending how wetting the liquid is (i.e., an effect of the contact angle). Once the shell forms, the only liquid surface area for evaporation would only consist of the interstices between the packed particles. In the second stage of drying, the liquid will have to be transported to the interstitial regions on particle-populated surface through capillary action.

From the above results, one can reasonably hypothesize the following: First, increasing initial particle concentration reduces the time to shell formation (t^*) decreases, resulting in larger sizes of the shell. On the other hand smaller initial concentrations allow more time for particle to redistribute themselves and lead to solid "microcrystal" [18] at the end of evaporation. Second, with smaller liquid–solid contact angles (θ), the effective surface area for evaporation is larger as the particle is more immersed in the liquid; smaller θ would thus allow evaporation to follow more closely the d^2-law. Third, particle mobility within the liquid would also affect their redistribution and thus the time to shell formation; high mobility allows particle to escape from early inclusion and thus longer t^*. Fourth, an increase in evaporation rate (K) should also decrease the time to shell formation, favoring hollow shell formation. Factors of the third and the fourth hypothesis can be combined into a non-dimensional Peclet number (Pe), the ratio of the evaporation and particle diffusion rate.

Two recent studies [20,21] investigated the evaporation characteristics of evaporation of nanofluid droplets containing various insoluble nanoparticles and several hydrocarbon liquids, respectively. It was found (a) that the value of K does not remain constant even before shell formation, (b) that K decreases with time and with decreasing droplet diameter, and (c) that the deviation from the d^2-law is more pronounced with higher initial particle concentrations. The authors of [20] thought the variation in K to be due to increases in an apparent enthalpy of vaporization with no clear physical explanation. In [21] it attributed the decreased evaporation rate to the particle aggregates within the droplet blocking the liquid pathway to the droplet surface. This study suggests that the deviation results from the particle aggregation inside the droplet, although such blocking is physically unlikely by aggregates residing inside the droplet. A more likely explanation should resemble the surface blockage by particle inclusion at the droplet surface described by Derkachov et al. [18]. Finding (c) appears to agree with the fact that higher initial concentrations should lead to earlier occurrence of inclusion and surface blockage, providing further support for explanation based on surface blockage.

One can find in literature models addressing various aspects of drying and evaporation of droplets containing particles having different sizes, not necessarily at nano-scales (see, for example [22–30]). This paper presents an analytical model for evaporation of droplets containing dilute particle suspension prior to shell formation, based on above findings (a) through (c) and surface blockage [16,18,20,21]. It is expected to address the hypothesis discussed above, with the following specific results: (i) that the model is capable of predicting critical phenomena such as the diameter (d^*) and the time at end of the first stage drying (t^*) and, reduction in the K value (i.e., departure from the d^2-law and how early it might occur during evaporation), and (ii) that these parameters can be explained based on the relative rates of liquid evaporation rate (K), particle size/liquid pair (particle mobility, D_{pl}) and their ratio, Peclet number (defined as $Pe \equiv K/8D_{pl}$) [31], the initial particle concentration (Y_{vo}) and the contact angle (θ). Evaporation of sessile droplet is not within the scope of this study (some results can be found in [32–34]).

2. The analytical model

Consider a single droplet containing nano-sized insoluble spherical particles that are initially evenly distributed within the droplet, as shown in Fig. 1 (phase (i)). As the liquid evaporates, some particles from the evaporated volume are "trapped" at the droplet surface ("inclusion" as described in [18] and phase (ii) in Fig. 1). These included particles are partly immersed in the liquid [11,35,36] and the exposed area of these included particles represent a reduction in the droplet surface from which liquid evaporate. The reduced (and effective) surface area for evaporation is denoted as A_e in Fig. 1, while the nominal surface of the droplet is A_t (the total area). Further evaporation and reduction of the droplet diameter leads to more included particles on the droplet surface. When the surface concentration of the particles reaches its saturated value at the droplet surface, a shell of densely packed particles forms at its surface [17] (phase (iii) in Fig. 1).

Therefore the shell is a layer of densely packed partially wetted particles. Since the particle size is far smaller than the liquid droplet size of engineering interest, the effect of curvature in the liquid–particle interfacial region is negligible [37]. The total of the interstitial area is thus A_e, which depends on the wettability and the contact angle of the given liquid–particle pair [37].

2.1. Effect of Peclet number on shell size

The particle motion depends on its diffusivity in the liquid, D_{pl}, which is calculated from Einstein–Stokes relation: $D_{pl} = k_B T/6\pi\eta r_p$, where k_B, T, η, and r_p are Boltzmann constant, temperature, liquid viscosity, and the particle radius, respectively. The relative motion of the regressing surface and the particles can be characterized by a dimensionless parameter, Peclet number (Pe) [17,38]. The characteristic time required for particles to diffuse across the droplet radius ($\tau_d = (d/2)^2/D_{pl}$) and the lifetime of a droplet is $\tau_e = d^2/K$, where d is the diameter of the droplet. Therefore,

$$Pe \equiv K/8D_{pl}$$

The scenarios for the extreme cases of $Pe \ll 1$ and $Pe \gg 1$ are shown in Fig. 2. For $Pe \ll 1$, the evaporation rate of a droplet is sufficiently low so that particles have sufficient time to redistribute themselves by diffusion throughout the droplet, resulting in a packed spherical particle aggregate. In this case, the size of final particle aggregate is determined by the initial number of particles, as would be for the microcrystal described in [18]. For $Pe \gg 1$, particles have insufficient time to diffuse and are quickly included on the regressing droplet surface, resulting in a hollow or doughnut grains [39,40]. For finite value of Pe, the shell diameter (d_s) depends