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Heat and mass transfer during levitation of a liquid nitrogen Leidenfrost droplet on a water pool

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Abstract. The understanding of cryogenic droplets evaporating on a liquid surface is of great interest in many industrial applications such as the Dearman engine, or food and beverage processing. However, due to the complexity of the physics involved, the entire process is far from being fully understood. Therefore, in the present study, the dynamics of a liquid nitrogen droplet deposited on a water surface is experimentally studied and theoretically modeled. During continuous cooling through the floating nitrogen droplet, the liquid at the pool surface may eventually solidify, which may significantly alters the situation for the evaporating droplet compared to the situation in which the pool remains liquid for the entire time. A crude theoretical model for the evaporation process of a floating nitrogen droplet is developed to calculate the vaporization rate of the droplet and the vapor layer thickness as a function of the radius of the droplet. The predictions from the model are compared to the experimental results, showing the lifetime predicted by the model is in good agreement with the experimental results.

1. Introduction

The evaporation of cryogenic liquid droplets on a liquid pool has a wide range of applications. For example, in the bottling industry, a liquid nitrogen droplet is dosed into the head space of a beverage bottle just before it is sealed [1]. Besides conservation of the beverage through a reduction of oxygen in the head space, the purpose of this nitrogen droplet during filling of plastic bottles is to allow the use of thin plastic bottles for packaging. Since the droplet expands to vapor whose volume is about 700 times larger than its liquid volume, the bottle is pressurized after closing providing the required stability of the bottle [2]. Liquid nitrogen droplets are also used in the Dearman engine where the rapid expansion of the droplets is transformed into mechanical work by mixing it with a heat exchange liquid which enhances the evaporation of liquid nitrogen inside the engine [3, 4]. However, very little is known about the evaporation process of a nitrogen droplet floating in contact with a warmer liquid, leaving the operators to adopt empirical or 'good feeling' approaches.

When a liquid droplet is placed on a surface whose temperature is much higher than the liquid boiling point, the liquid droplet will be in the film boiling regime, i.e. it levitates on a layer of its own vapor which gives the droplet very high mobility [5]. This phenomenon is referred to as the Leidenfrost effect [6] and accordingly, such floating droplets are referred to as Leidenfrost droplets [7]. The vapor film acts thermally insulator resulting in a severe reduction of the heat transfer rate between the droplet and the pool compared with a droplet being in direct contact with the surface.

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CEC 2021		IOP Publishing
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A number of researchers have studied Leidenfrost droplets evaporating on a hot substrate, both theoretically and experimentally [7–11]. Gottfried et al. [7] proposed the first theoretical model in 1966 to describe small droplets evaporating on a hot substrate, which was validated by means of experiments showing less than 10 % deviation in terms of the lifetime of the droplet. A measurement technique for determining the vapor layer thickness below a Leidenfrost drop on a solid surface was developed and applied by Burton et al. [8]. Sobac et al. [9] proposed a model for the vapor layer thickness on a solid surface, which was in agreement with the experimental results from Burton et al. In the model, the Navier-Stokes equations are solved for the vapor layer under the assumption of a lubrication flow. The pressure in the vapor layer results from both the hydrostatic pressure and Laplace pressure which is caused by the curvature of the drop, therefore making a relation to the shape of the drop. Maquet et al. [10] adapted this model to liquid surfaces integrating the deformability of the liquid meniscus. Van Limbeek et al. [11] further improved this model by a matched asymptotic analysis in the case of low evaporation numbers. However, due to the inherent complexity of liquid nitrogen vaporization on water, no complete description has yet been reported in the literature.

In the present case of a nitrogen droplet evaporating on a water surface, the water surface is exposed to a fluid being at a much lower temperature than the freezing temperature of water. Consequently, a decrease of the water surface temperature below the pool freezing temperature may alter the situation for the evaporating droplet when ice forms in the pool below. It is of great importance both from scientific and industrial perspective to understand the physical processes involved. Therefore, the present work focuses on elucidating the evaporation process of a liquid nitrogen droplet floating on a water pool by proposing a crude theoretical model to predict the lifetime of the droplet and performing experiments to validate this theoretical model.

2. Theoretical Model

The main objective of the proposed theoretical model is the prediction of the lifetime of a nitrogen droplet through a theoretical description of the physical processes involved. In that scope, the liquid nitrogen droplet is approximated as a cylinder, as schematically pictured in Figure 1, which hopefully gives the same scaling law as the spherical case.



Figure 1: Schematic illustration of the situation during floating of a nitrogen droplet on a water pool.

The liquid nitrogen droplet with a height of 2a and radius a in a cylindrical shape is assumed to float on a flat water surface. The water pool which may eventually comprise ice formed below the nitrogen drop is assumed uniform at the melting point of the water pool, 273.15 K and the

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liquid nitrogen drop can be assumed at saturation temperature. For simplification purposes, the height of the vapor layer δ is assumed uniform along the radial direction. It is essential to understand the dynamics of the vapor layer to predict the heat flux from the surface to the liquid and finally determine the rate of droplet evaporation. Conduction through the vapor layer is generally the main heat exchange mechanism between the liquid and the surface [12]. Thus, only conductive heat transfer from the pool to the bottom of the droplet is considered in the model while all other sides of the droplet are considered as adiabatic. Under lubrication approximation, i.e., when the scale of the cylinder diameter 2a is much larger than δ , such as in the present case, the Navier-Stokes equation reduces to

$$\mu^{-1}\frac{\partial p}{\partial r} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u_r}{\partial r}\right) - \frac{u_r}{r^2} + \frac{\partial^2 u_r}{\partial z^2},\tag{1}$$

where μ is the viscosity, u_r the velocity in radial direction, p the pressure, and z and r are the axial and radial directions, respectively. In order to derive the velocity field in the vapor layer, the radial vapor velocity u_r is assumed as

$$u_r(r,z) = \overline{u}_r f(z), \tag{2}$$

in which $\overline{u}_r = \frac{1}{\delta} \int_{-\delta/2}^{\delta/2} u_r(r, z) dz$ is the height averaged velocity variable along the *r*-direction. Assuming a constant flux of evaporating nitrogen at the bottom of the droplet, from conservation of mass it follows

$$2\pi r \delta \overline{u}_r \rho_v = \frac{\pi r^2}{\pi a^2} \dot{m},\tag{3}$$

where ρ_v is the density of the vapor layer, and \dot{m} the total mass flow rate of the vapor released underneath the liquid nitrogen droplet. By using equations (2) and (1), the momentum balance, equation (1), reduces to

$$\frac{\partial p}{\partial r} = \mu \frac{\partial^2 u_r}{\partial z^2} = \mu \overline{u}_r \frac{\partial^2 f(z)}{\partial z^2} \tag{4}$$

and rearranging leads to

$$(\mu \overline{u}_r)^{-1} \frac{\partial p}{\partial r} = \frac{\partial^2 f(z)}{\partial z^2}.$$
(5)

It is obvious that the only solution for equation (5) is that the left side and the right side of the equation is equal to constant. Therefore, the solution for f(z) is found as

$$u_r(r,z) = \frac{3}{2}\overline{u}_r(1 - \frac{4z^2}{\delta^2}).$$
 (6)

Using equation (3) into equation (4), the pressure field is derived as

$$p(r) = \frac{3\mu r\dot{m}}{\pi\rho_v \delta^3} (1 - \frac{r^2}{a^2}) + P_o.$$
 (7)

Balancing the mass of the liquid nitrogen droplet with the force acting on it through the pressure resulting from the vapor flow yields

$$2\pi a^3 \rho_n g - \int_0^a p(r) 2\pi r \,\mathrm{d}r = 0, \tag{8}$$

where ρ_n is the density of liquid nitrogen. Finally, from using equation (7) in equation (8), the vapor layer thickness is found as

$$\delta = \left(\frac{3}{4} \frac{\mu \dot{m}}{\pi \rho_v \rho_n g a}\right)^{1/3},\tag{9}$$

IOP Conf. Series: Materials Science and Engineering 1240 (2022) 012157

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still depending on the unknown evaporative mass flux, \dot{m} , representing that physical quantity coupling the fluid flow problem with the heat transfer problem. By applying Fourier's law, the heat flux is given as

$$q'' = -k\frac{\mathrm{d}T}{\mathrm{d}x} = -\frac{k_n + k_b}{2}\frac{T_n - T_b}{\delta},\tag{10}$$

where T_b is the water/ice surface temperature, and T_n is the saturation temperature of liquid nitrogen. This heat flux is related to the evaporating total mass flow rate through the latent heat, L_{eff} , as

$$q'' = L_{eff}\dot{m}.\tag{11}$$

Here, L_{eff} refers to the effective latent heat which incorporates both the latent heat required to evaporate the liquid nitrogen droplet and the sensible heat consumed to warm up the nitrogen vapor to the temperature it has when the vapor leaves the gap, which will be above the saturation temperature of nitrogen. Then the effective latent heat is expressed as

$$L_{eff} = L + (c_{p,ave}T_{ave} - c_{p,n}T_n), \tag{12}$$

where $c_{p,ave}$ and $c_{p,n}$ denote the heat capacity at the average vapor temperature in the gap, $T_{ave} = (T_n + T_b)/2$, and T_n , respectively [13].

With equations (10)-(12), the evaporative mass flow of nitrogen vapor can be expressed as

$$\dot{m} = 2\rho_n \pi \frac{\mathrm{d}a^3}{\mathrm{d}t} = \frac{q''}{L_{eff}} = -\pi a^2 \frac{k_n + k_b}{2L_{eff}} \frac{T_n - T_b}{\delta},\tag{13}$$

and by using equations (13) and (9), the vapor layer thickness is found as

$$\delta = \left(\frac{3\mu}{4\rho_n \rho_v g} \frac{k_n + k_b}{2L_{eff}} (T_b - T_n)\right)^{1/4} a^{1/4}.$$
 (14)

Finally, the temporal evolution of the droplet radius is obtained as

$$a^{5/4} = a_0^{5/4} - Ct, (15)$$

in which

$$C = \frac{5(k_n + k_b)}{48\rho_n L_{eff}} \frac{(T_b - T_n)}{\delta^*},$$
(16)

where $\delta^* = \left(\frac{3\mu}{4\rho_n\rho_v g}\frac{k_n+k_b}{2L_{eff}}(T_b-T_n)\right)^{1/4}$. Therefore, the lifetime of a nitrogen droplet can be predicted as a function of the initial radius of the droplet from equation (15) as

$$\tau_{tm} = a_0^{5/4} / C. \tag{17}$$

3. Experimental Setup and Measurement Methods

The theoretical model is validated using data obtained with the experimental setup schematically shown in Fig 2. In order to determine the behavior of the nitrogen drop and the ice which forms under the meniscus, two high-speed cameras (Phantom VEO 710 L) are used for capturing the situation from the top and from the side. Two LED-arrays are used to illuminate the drop and meniscus from below, resulting in blacklight shadowgraphy videos when filmed with a high-speed camera from above through a mirror. The lenses used for the top view and side view observation are a Laowa 25 mm f/2.8 2.5-5x Ultra Macro, and a Navitar 12x zoom lens, respectively.

IOP Conf. Series: Materials Science and Engineering 1240 (2022) 012157 doi:10.1088/1757-899X/1240/1/012157



Figure 2: A sketch of the experimental setup

The water in the cell is de-ionized Milli-q water and the meniscus suspended in the lid of the cell has a radius of 5.70 mm. To perform the experiments, a liquid nitrogen droplet is first placed on a spoon. Once it is reduced to the desired size by evaporation, it is rolled onto the water meniscus over a ramp which gives more control for positioning of the droplet on the meniscus. Immediately after the droplet makes contact with the pool, the cameras are triggered to capture the process. From the captured high-speed top-view videos, the temporal evolution of the size of the nitrogen droplet is obtained from video post-processing using an in-house code written in Matlab[®]. Example images extracted from the videos captured in the top and side view are shown in Figure 3 (a) and (b), respectively.



Figure 3: Example images captured during an experiment, showing (a): Top-view image of a nitrogen drop together with (b): Side-view image of the ice cap forming in the water pool below a nitrogen droplet. Note that due to total internal reflection at the liquid/gas interface, the meniscus appears completely black and the nitrogen droplet is not visible in (b).

4. Results and Discussion

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Experimental results for the temporal evolution of the nitrogen droplet radius is shown in Figure 4. While the pure data of the temporal evolution of the radius of evaporating droplets determined from several repeated experiments is shown in Figure 4 (a), the experimental data normalized with the initial diameter and total evaporation time of the individual droplets is shown in Figure 4 (b), revealing that for that normalization all lines collapse and show a linear trend confirming the experimental results observed from Maquet et al. [10]. Figure 4 (c) shows the normalized radius to the power of 5/4 together with the theoretical prediction. The reason for the slightly disagreement between the experiment and the theoretical model will be discussed in this session.



Figure 4: (Color online) Radius of the nitrogen droplet as a function of time showing (a): Temporal evolution of the nitrogen droplet radius obtained from the experiments, (b): The radius and time normalized with the initial radius and the total lifetime of the droplets in which blue line is a linear guide line, respectively, and (c): The normalized radius raised to the power of 5/4 together with the theoretical prediction. Note that the experimentally determined total lifetime has been used for normalization of time in (b), while the theoretical values have been used in (c).

A comparison between the predicted lifetime and experimental results is shown in Figure 5 (a). The lifetime predicted using the theoretical model is a bit longer than experiments as shown in the picture. The deviation may result from the constant water/ice temperature assumption in the model. In reality, the water/ice surface temperature continues to decrease as the ice grows instead of remaining at the freezing point of water. Assuming the water/ice surface at the

IOP Conf. Series: Materials Science and Engineering

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Figure 5: Comparison of predictions from the theoretical model with experimental results showing (**a**): The predicted lifetime of the nitrogen droplets, and (**b**): The theoretical thickness of the vapor layer as a function of the droplet size compared with one experimental result from Chandra [14].

melting point of water is valid if the temperature difference in the vapor layer is much higher than that in the ice. The thermal conductivity of ice is approximately 140 times larger than that of the nitrogen vapor, and based on the experimental results the ice thickness is around 0.1 mm. By using equation (10), the temperature difference of the ice from bottom to the top is found in the order of 5 K. However, the temperature difference in the vapor layer is the difference between the freezing temperature of the pool and the saturation temperature of liquid nitrogen which is around 195 K. Thus, assuming a constant water/ice temperature should not impose a major effect on the model. Another possible source for errors of the model may be the prediction of the vapor layer thickness. Figure 5 (b) shows the theoretical prediction for the vapor layer thickness depending on the droplet size. Since experimental data on the vapor layer thickness below a nitrogen Leidenfrost droplet is almost not available in the literature, the theoretical data is compared with an experimental result from Chandra et al. [14] for the vapor layer thickness below a nitrogen droplet floating on a hot glass substrate. As shown in Figure 5 (b), for a droplet size of 1 mm, such as in the mentioned experiment from Chandra et al. [14], the vapor layer thickness is well predicted using the theoretical model.

As shown before, the present experimental results on the evolution of the droplet diameter confirm the results reported in Maquet et al. [10], that the droplet diameter decreases linearly with time. However, while the lifetime is slightly overestimated with the model, the vapor layer thickness is slightly underestimated using the model. One of the main reasons causing the inaccuracy may result from the strongly simplified shape of the droplet and the pool surface. Another reason is that only the heat flux from the bottom of the liquid nitrogen droplet is considered and other sides of the droplet are assumed to be adiabatic which leads to a longer lifetime predicted by the theoretical model.

5. Conclusion

In the present study, a theoretical model has been proposed to describe the evaporation process of a liquid nitrogen droplet floating on a water pool. In the model, the droplet shape is cylindrical with a uniform vapor layer thickness established between the droplet and the meniscus. The gas flow below the droplet is obtained under lubrication approximation. This assumption is justified as only the conductive heat transfer from the pool to the bottom of the droplet is considered.

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IOP Conf. Series: Materials Science and Engineering	1240 (2022) 012157	doi:10.1088/1757-899X/1240/1/012157

The temporal evolution of the vapor layer thickness and radius of the droplet are predicted by the model. Experiments have been performed in order to validate the model, finally showing a good agreement between the experimental and theoretical results in terms of the temporal evolution of the droplet size. However, the present model still needs to be improved by e.g. considering the effect of the ice formation and the influence of heat flux from other sides of the droplet than only the bottom.

6. Appendices

Variable	Unit	Quantity	Comment
$\overline{T_n}$	K	77.3	
T_b	Κ	273.15	
T_{ave}	Κ	175.23	
P_o	MPa	0.101325	Atmospheric pressure
μ	$\mu Pa \cdot s$	11.5	Taken at T_{ave}, P_o
$ ho_v$	$ m kg\cdot m^{-3}$	1.96	Taken at T_{ave}, P_o
$c_{p,ave}$	$kJ \cdot kg^{-1} \cdot K^{-1}$	1.05	Taken at T_{ave}, P_o
$c_{p,n}$	$\mathrm{kJ}\cdot\mathrm{kg}^{-1}\cdot\mathrm{K}^{-1}$	1.12	Taken at T_n, P_o
$ ho_n$	$ m kg\cdot m^{-3}$	806	Taken at T_n , P_o
g	$\rm m\cdot s^{-2}$	9.81	
k_n	$\mathrm{mW} \cdot \mathrm{m}^{-1} \cdot \mathrm{K}^{-1}$	7.2	Taken at T_n, P_o
k_b	$\mathrm{mW} \cdot \mathrm{m}^{-1} \cdot \mathrm{K}^{-1}$	24.0	Taken at T_b , P_o
L_{eff}	${ m kJ\cdot kg^{-1}}$	295	

Table 1: Values used for calculation (the values are taken from REFPROP.)

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