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Deposition of metallic colloids under sub-cooled nucleate boiling

Hitesh Bindra[a,*], Barclay G. Jones[b]

[a] Levich Institute, City college of New York, New York, NY 10031, United States
[b] Department of Nuclear Plasma and Radiological Engineering, University of Illinois at Urbana Champaign, Urbana 61801, United States

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ABSTRACT

Heterogeneous nucleate boiling causes the deposition of colloidal particles and governs the growth rate of porous deposit layers on heating surfaces when dilute metallic colloidal suspensions are present in aqueous medium. Post-experimental analysis reveals that these deposits are found in the form of spots of variable thickness around the bubble nucleation site. It is shown here that thickness of these spots and deposit amount is directly related to evaporation rate which is dependent on heating wall temperature and fluid temperature. The experimental observations suggest that the deposition occurs around the contact line of bubble and extends underneath area of bubble micro-layer. Rate of transportation of particles to the wall is directly related to frequency of bubble nucleation and departure diameter of departing bubbles. These transported particles deposit onto the heating surface with an attachment probability, which is modeled here as a function of colloid–surface interaction potential. A model of deposition rate has been developed on the basis of evaporative flux E_q'', attachment probability p_{att} and bulk concentration of colloidal particles C_0. This model provides qualitative explanation for experimental results and suggests that deposition flux has linear dependence on the quantity $p_{att}C_0E_q''$.

1. Introduction

Heterogeneous nucleate boiling is the most common form of boiling observed in real life, with applications ranging from boiling water in a faucet to steam generators in power plants. It results in the deposition of impurities such as particulates, salts, and colloids, etc., which are present as a suspension or solute form in the heat transfer fluid, on the heating surface. Various boiling experiments [1,2] with suspended nano-particles showed that these particles form layered deposits on the heated surface which have an improvised impact on surface wettability. The reason attributing to this increase in surface wettability might be higher surface roughness for the deposit layers compared to the bare surface, leading to Wenzel state. Despite research efforts of a decade, it is still unknown how to control this surface roughness by deposition from these boiling experiments so that surface wettability can be tuned. In the real world, industrial scale boilers and nuclear power plants have experienced the deposition of corrosion products on the heat exchange surfaces and the adverse consequences associated with it such as CRUD induced power shift in pressurized water reactor (PWR) type of nuclear power plants [3]. The eroded corrosion products in the bulk stream comprise of metal oxides (size order ∼10 nm to ∼10 μm) in the form of dilute suspensions, which can be defined as colloidal suspensions. The most common examples of such dilute colloidal suspensions are found in process industries with carbon steel piping, wherein the corrosion products mostly constitute of Fe_2O_3 present in water in very dilute quantities (10 ppb to 1 ppm). The post examination of industrial scale heat exchange surfaces showed the formation of irregular, non-uniform and thick porous layers (100–300 μm). Whereas, laboratory experiments conducted for time scales such as minutes or hours show that these particles deposit selectively at nucleation sites on the heated surface in the form of circular discs and spots. It is difficult to correlate the deposition data from such laboratory experiments with the deposits on steam generators, evaporators, and nuclear fuel surfaces which operate for several months. Therefore, it is essential to understand the basic mechanism for this boiling assisted deposition and develop phenomenological models. Large scale laboratory experiments [4–6] were useful in formulating empirical relations between overall deposition rate of porous layers and operating flow, pressure, heat flux and concentration of the particles. Still, the basic mechanism of boiling-assisted deposition of colloids is unclear. The few consistent mechanistic observations of the prior experimental work are; (a) deposits are in the form of porous layers (b) experiments on small time scale such as minutes or hours show that deposits are in the form of isolated spots or rings representing active bubble nucleation sites. Pool boiling [7] and flow boiling [8] experiments with colloidal suspensions showed that spot-like circular deposit shapes obtained were similar to deposit shapes obtained with boiling of insoluble salt solutions

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* Corresponding author. Tel.: +1 212 650 6720; fax: +1 212 650 6660.
E-mail address: hbindra@che.ccny.cuny.edu (H. Bindra).

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Nomenclature

d_b	bubble departure diameter (mm)
f	frequency of bubble nucleation (1/s)
n_c	no. of nucleation sites
f_c	frequency of bubble nucleation at each site (1/s)
h_{lv}	latent heat of vaporization (J/kg)
C_p	specific heat (J/kg-K)
ρ_l	liquid density (kg/m^3)
ρ_v	vapor density (kg/m^3)
V_{vdw}	Van der Waals potential energy (kT)
V_{elec}	electrostatic potential energy (kT)
V_t	total interaction potential energy (kT)
ε	vacuum permittivity (8.85×10^{-12} F m^{-1})
ε_0	relative permittivity of water (55.8 at 100° C)
r_p	radius of particle (nm)
e	electron charge (1.6×10^{-19} C)
k	Boltzmann constant (1.38×10^{-23} J/K)
σ	surface charge density (C/m^2)
G	free energy (J)
ψ	zeta potential (mV)
n_0	concentration of ions (M)
C_0	bulk colloidal concentration (ppm(w/w))
T	temperature (°C)
T_w	wall temperature (°C)
T_s	saturation temperature (°C)
$T_\propto$	bulk temperature (°C)
κ	Debye radius inverse (nm^{-1})
K_a	attachment rate coefficient (m/s)
p_{att}	attachment probability
E_q	evaporation rate (mm^3/s)
E_q''	evaporation flux (mm^3/cm^2-s)
ρ_p	density of particle (gm/cm^3)
D	diffusivity of colloid (cm^2/s)
ν	kinematic viscosity (cm^2/s)
δ_f	interaction force boundary layer thickness (nm)
x_{cp}	distance of closest approach (nm)

[9]. The fundamental studies of vapor bubble growth during boiling from hot surface suggest that there is a liquid layer present, having thickness of few microns, between vapor bubble and heated surfaces. So the actual contact area of the vapor bubble is very small for surfaces such as metals or metal oxides. It was proposed that drying out of this liquid layer beneath the bubble leaving suspended particles on the surface might be the mechanism of deposition [8]. However, this proposed model did not explain the observed effect of chemistry on attachment or re-entrainment of particles. The bubbles in these experiments had very small residence times (*O*–10 ms) and no visual data on micro-layer evolution or dry-out underneath the bubbles is available yet. In a similar but more controlled experimental environment known as 'coffee-ring' experiment, more consistent theory and easily reproducible data were provided. Quantitative modeling [10] proposed to predict rate of growth of deposit based on evaporation flux around drop contact line matched very well with deposition profile and data. Similar experiments [11] with evaporating droplet of colloidal suspension with DLVO (Derjaguin–Landau–Verwey–Overbeek) interactions showed significant impact on deposition pattern. The evaporation rate for such experiments is much slower and for droplets evaporating at faster rate the contact line does not remain pinned necessarily and makes jumps causing dispersed deposits [12]. This can be expected under pool boiling conditions as well where wall superheats make the evaporation process much faster than 'coffee-ring'. Some recent reports with high fidelity experimental techniques have been helpful in determining contact line evolution during vapor bubble growth on a heated surface [13]. The limited choices of materials, which are essential to observe attachment kinetics, during experimental simulation, make it impossible to have such detailed observations during colloidal deposition experiments. However, in sub-cooled boiling the bubble growth time is much faster than waiting for another bubble to appear. Therefore the frequency of bubble generation and departure at multiple sites determines the rate of evaporation on the heated surface rather than growth and evolution of a single bubble. In this work the deposition characteristics viz. rate, size and thickness of these deposits dependent upon boiling parameters i.e. bubble frequency and departure diameter have been examined experimentally and a model is formulated based on phenomenological understanding. The motivation for this work was to generate boiling assisted colloidal deposition data and model, excluding the effects from forced circulation and complicated water chemistry.

2. Experimental method

Various tests are conducted at different wall temperatures and sub-cooling levels in quiescent pool conditions. A large cuboidal Plexi-glass tank is chosen to house the heater and test piece in such a way that the boiling can be observed on a small metal test-piece in a large aqueous medium. It is desired that heater test piece set up has reduced impact from flow disturbances and to provide a virtually infinite pool of dilute solution of metallic colloids (10 ppm). Due to such large pool of colloidal assembly, change in concentration levels is negligible during the experiments which are conducted for 2 h each. Experimental set-up is shown via a diagram in Fig. 1. Temperature of the bulk liquid in the tank can be independently controlled using immersion heaters at two ends of the tank to avoid thermal gradients in particular directions, thus maintaining uniform bulk temperature. Pressure of the system is atmospheric and corresponding saturation temperature is 100 °C; bulk tank temperature $T_\propto$ is maintained at constant value with operation range from 85 °C to 97 °C. Arrangement is set to have an access of high-speed visualization of bubble dynamics with CCD camera. Heating wire made of nichrome is wrapped around grooves on the anodized aluminum hollow cylinder which is adhered to the solid copper block with ceramic paste. This provides a 100 W (30 W/cm^2 on the boiling surface) maximum power rating of the heater with maximum current of 4.5 A. The whole heater assembly is insulated in radial direction and heat is conducted in axial direction to heat the liquid. Heat is conducted through the copper block to the test piece through liquid metal joint to provide uniform heat flux over 1 cm^2 area of the test piece. Wall temperature and bulk temperature, important parameters to directly impact boiling process, are measured via thermocouples. K-type thermocouples are employed having accuracy of ±1.1 °C to help in monitoring feedback for variable resistance power supplies. High thermal conductivity of test piece avoids wall response time lag during bubble growth process.

Particles tested in the system are hematite due to their practical importance as corrosion product. Therefore, alumina having high thermal conductivity and with different iso-electric point (IEP) from hematite is selected as the test piece. IEPs for the surface ($Al_2O_3 \sim 9.0$) and particles ($Fe_2O_3 \sim 6.6$) allow low attachment resistance if the pH of bulk solution is in between, in these tests it is 7.5. Average roughness of test piece after surface treatment is less than 0.1 μm, measured with DEKTAK stylus profilometer. Nominal spherical particle size after sonication and dilute solution preparation is ~34 nm for most of the tests. Particle size and zeta-potential values were recorded for each test performed with