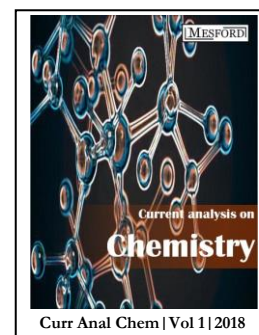


Evaluations of Surfactant Solutions for Nucleate Pool Boiling at Low Heat Fluxes

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Abstract:

In this study, nucleate pool boiling of surfactant solutions at low heat fluxes is investigated. The surfactants chosen for the study are an ionic sodium lauryl sulfate (SLS), nonionic ECOSURF™ EH-14, and nonionic ECOSURF™ SA-9. It is observed that adding a small amount of surfactant alters the water boiling phenomenon considerably. Boiling curves for different concentrations are shifted to the left. The wall temperature dropped with an increase in the concentration of aqueous surfactant solutions. Also, it is found that the boiling heat transfer enhancement of SLS is higher than that of EH-14 and SA-9 compared to water. Boiling heat transfer coefficient (h) enhancements compared to water are 46%, 30%, and 21% (for SLS, for EH-14 and for SA-9 respectively).

Results prove that there is an important possibility to enhance the boiling application processes by environmentally friendly EH-14, and SA-9 additives. Experimentation can be extended for searching other surfactants in order to find their most efficient quantity in water for boiling heat transfer.

Publication History: Received: 06 August 2018 | Revised: 31 August 2018 | Accepted: 21 September 2018

Keywords:

Boiling, heat transfer, surfactants, surfactant solutions, pool boiling, phase change.

1. INTRODUCTION

Boiling is the most effective heat transfer method because of its high performance due to latent heat. Therefore, boiling allows to reduce size, weight and volume of heat exchange devices and to improve the thermal performance of components for the process industry and power plants. It has been found in a wide range of applications in both traditional industries such as various energy conversion systems, heat exchange systems, air-conditioning, refrigeration systems, and in highly specialized fields such as cooling of high energy-density electronic components, micro-fabricated fluidic systems, and the thermal control of aerospace stations [1]. Steam generators can be better designed if the boiling process is known in detail. This would improve the thermal cycle and the plant efficiency [2].

The reasons for using the boiling process vary. In a power station using steam turbines, the vapor itself is the desired product. When cooling electronic components, it is the high heat transfer characteristics of boiling that are important. On the other hand, in the analysis of coolant accidents in water-cooled nuclear power stations, it is the rather poorer heat transfer that can occur at high surface temperatures that is of interest [3]. When the temperature of the heated surface gets

very high, the characteristics of the boiling process change (Critical Heat Flux) and it is no longer possible for the liquid to come into good contact with the surface. A film of vapor separating solid and liquid reduces heat transfer [3].

An extensive literature exists on methods of enhancing liquid-solid heat transfer rates, which allow the size, cost, and complexity of such equipment to be reduced. One promising approach has been the use of additives that enhance heat transfer by altering liquid properties: addition of surfactants to boiling water is known to improve heat transfer [4]. Adding surfactants can lower the liquid-vapor surface tension and increase the bubble departure frequency, thereby enhancing heat transfer [5]. Surfactant compositions have unique physical properties that will significantly affect the boiling and evaporation behavior.

Heat transfer rates depend not only on the physical properties of the liquid, but also on the method by which it is supplied to a solid surface. Water sprays are widely used in applications such as cooling of hot surfaces in the metallurgical industry and fire extinguishment by sprinkler systems. Addition of a wetting agent considerably reduces the volume of water required to extinguish fires [4].

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Boiling of binary and ternary mixtures have also been found to change the boiling behavior due to the liquid-liquid molecular forces and solid-liquid and vapor-liquid interactions such as surface tension. Experimental studies on enhancing the pool boiling heat transfer coefficient of binary dilute mixtures of water/glycerol, water/MEG (Mono-ethylene glycol) and water/DEG (di-ethylene glycol) have been carried out by Sarafraz et al. [6]. Results showed that presence of ammonium salts into the mixtures lowers the surface temperature and caused higher pool boiling heat transfer coefficient [6].

In other study, to quantify the forced convective and nucleate flow boiling heat transfer coefficient, Sarafraz et al. performed experiments with Al₂O₃ water based nanofluid [7]. With increasing the heat and mass flux, heat transfer coefficient increased dramatically [7]. Flow boiling heat transfer coefficients of deionized water and copper oxide water-based nanofluids at different operating conditions have also been experimentally measured by Sarafraz et al. [8]. Flow boiling heat transfer coefficient increased with increasing the applied heat flux [8].

Experimental investigations on pool boiling heat transfer to functionalized and nonfunctionalized carbon nanotube nanofluids have been performed on a discoid heater under the atmospheric pressure [9]. As a result, functionalized carbon nanotube was suggested a promising option for nucleate boiling heat transfer, since it enhanced the heat transfer coefficient and critical heat flux [9]. In another study on the thermal performance of a copper-made heat sink with rectangular microchannel, Carbon nanotube (CNT) aqueous nanofluid is used as a coolant inside the microchannel [10]. CNT aqueous nanofluids showed approximately 29% enhancement in heat transfer coefficient over the base fluid. [10].

2. BOILING WITH SURFACTANTS

A number of studies have been performed to investigate boiling phenomena with surfactants including both nucleate pool boiling and flow boiling.

Considering the role of surface tension in boiling heat transfer, Westwater [1] assumed the following heat transfer coefficient relationship with surface tension:

$$h \propto \sigma^n \quad (1)$$

Literature is contradictory about the role of surface tension during boiling process. Some researchers have reported that surface active agents in water increase heat transfer at a given temperature difference driving force, while other researchers reported a decrease. Different values have been published for the exponent n , which has values of -2.5, -2, -1, +0.25, +1.275 [1]. This is conflicting for the role of surface tension in the boiling process. However, theoretically surface tension is an important variable in boiling process. Rate of formation of vapor nuclei in the boiling of a liquid is proportional to surface tension as [1]:

$$N \propto e^{-\sigma^3} \quad (2)$$

Therefore, small decrease in surface tension should cause big increase in the number of nuclei. This has been observed by the nucleate boiling process conducted by many researchers. For example, Zhang [11] observed the nucleate boiling process by means of a high-speed camera and compared the observed results for water and surfactant solutions. His observation has confirmed this point. In addition, cavitation theory predicts that force required to rupture a liquid in tension is proportional to surface tension as [1]:

$$F \propto \sigma^{3/2} \quad (3)$$

Therefore, liquids with large surface tensions should be difficult to fracture.

The nucleate pool boiling heat transfer has generally been observed to increase with increasing the concentration of aqueous surfactant solutions. However, when the solution concentration is larger than critical micelle concentration (cmc), there will be reduction in boiling heat transfer enhancement [12].

It is generally concluded that the enhancement of saturated nucleate pool boiling heat transfer by the addition of small amounts of surfactants in water is because of the reduction of surface tension. However, some researchers found different results. Lin *et al.* [13] reported that some surfactants could enhance saturated nucleate pool boiling heat transfer while some did not. Yang *et al.* [14] studied saturated nucleate pool boiling of water with surfactants (Triton SP-190 and Triton SP-75) on a cylindrical surface. They reported that while both equilibrium surface tension and contact angle were reduced, the nucleate pool boiling heat transfer was hardly affected. The possible reason could be the conflicting functions of surface tension and contact angle. The reason why some surfactants can reduce surface tension of aqueous surfactant solutions but cannot enhance nucleate boiling heat transfer is not clear. The possible reason could be the effect of surfactant type [1].

3. EXPERIMENTAL TEST SETUP

The experimental setup is designed to provide controlled, repeatable boiling conditions for surfactant solutions. Fig. (1) shows the schematic diagram of the experimental apparatus.

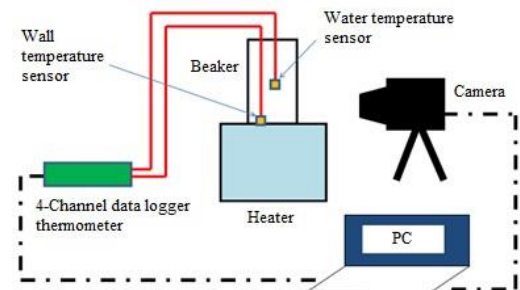


Fig. (1). Schematic of experimental test setup [27].

The experimental apparatus consists of the following main components: beaker, hot plate, thermometer, thermocouples, image acquisition, and precision scales.

A data acquisition PC collects and displays temperature sensor readings and includes a large stopwatch display for clear experiment coordination and logging. The RDXL4SD model of temperature recorder is used. Therefore, the data logger is able to collect temperature data of four channels with the time information and saves them in an Excel file, which can be moved to a computer for analysis. Type K thermocouples were used with adhesive mounts to ensure measurement on the beaker's interior bottom surface and water temperatures. This thermocouple has ultra-slim silicone rubber, which provides high flexibility, and it is capable to resist a variety of chemicals and oils. Also, it has a self-adhesive foil backing for faster response time. Response time of thermocouples is 0.002 sec (in still H₂O). Tolerance value for K-type thermocouple is $\pm (0.4\% + 1^\circ\text{C})$. The graduated transparent beaker enables clear viewing for the digital camera and enables observation of the amount of liquid evaporated. Surfactant is added into 400 mL of water and mixed for 1 minute with magnetic stirrer unit in the hot plate. A magnetic stirrer agitates the water by using a stir bar inside the hot plate. After stirring, the mixture is heated with the hot plate. Image acquisition is employed to observe and report the mechanisms of pool boiling heat transfer of surfactant solutions. A camera type CASIO EX-FH-20, was used to record pool boiling phenomena and bubble dynamics. The camera can record up to 1000 frames per second of video and shoot continuous high-resolution images at speed 40 images per second. To reach the uniform heat flux, the temperature distribution of the hot plate was observed with Infrared camera. The beaker is positioned on the same uniform temperature area for all tests.

Anionic surfactant (SLS) and nonionic surfactants (EH-14 and SA-9) are tested for various concentrations. Properties of the surfactants are given in Table 1. Surface tension measurements of varying concentrations (ppm) of aqueous SLS, SA-9, EH-14 solutions are studied in other studies of Dikici *et al.* [15-16]. The aqueous solutions of surfactants are prepared by dissolving the measured samples of surfactants in water. The amount of surfactant was measured by using the precision scale depending on concentration. Parts per million unit is the mass ratio between the surfactant and the solution, and ppm is defined as:

$$ppm = 1,000,000 m_{surf} / m_{sol} \quad (4)$$

For each value of heat flux, the collected data included the wall temperature, bulk water temperature, and time to reach boiling point for all tests. Also, the digital camera was used to record and capture images of the boiling process (bubble nucleation, growth, and departure). Then, the heat flux was changed, and the same procedure was repeated after test setup was cooled to room temperature.

NDJ-5S digital rotary viscometer was used to determine the viscosities of three surfactant solutions of SLS, EH-14, and SA-9 of various compositions at room temperature.

Table 1. Physico-Chemical Properties of Surfactants [27].

Properties	SLS	ECOSURFTM EH-14 (90% Actives)	ECOSURFTM SA-9
Chemical description	CH ₃ -(CH ₂) ₁₁ -O SO ₃ ⁻ -Na ⁺	Alcohol Alkoxyate	Seed oil surfactant
Surfactant type	Anionic (-)	nonionic	nonionic
Molecular weight (g/mol)	288.372	1036	668
Surface Tension (mN/m)	38.0	31.8	29
Appearance	White powder (Solid)	Clear slippery liquid	Pale yellow liquid
Cloud point (°C)	-	86	57
Solubility	Soluble in water	Soluble in water	Dispersible in water

Sodium lauryl sulfate (SLS) is an anionic surfactant used in many cleaning and hygiene products. ECOSURF™ EH-14 surfactant is biodegradable, nonionic surfactant with low aquatic toxicity. ECOSURFTM SA-9, known as a seed oil surfactant, is also a biodegradable nonionic surfactant that composed of alcohols, C₆-C₁₂, ethoxylated, and propoxylated 55-80%.

4. GOVERNING EQUATIONS

Boiling is subcooled when the temperature of the liquid is below the saturation temperature, T_{sat}. During subcooled boiling, the thermal energy from the hot plate is transferred to the water as sensible heat. This heat is used to raise the temperature of water from its initial temperature, T_i to the saturation temperature T_{sat}. [17]. In Eqn. (5) q_{in} is the net heat transfer from the hot plate to the water.

$$q_{subcooled}'' = \frac{m C_p \Delta T}{A \Delta t} \quad (5)$$

Boiling is saturated when the temperature of the liquid is equal to the saturation temperature, T_{sat}. The heat from the hot plate in this stage is transferred to the boiling water as latent heat. All of the heat transferred to the boiling water is used for a phase change from liquid to vapor. Eqn. (6) first relates the heat from the hot plate, q_s, to the convective heat transfer to the water. It then equates the same heat, q_s, to the heat escaping during boiling mass transfer.

$$q_{subcooled}'' = h(T_s - T_{sat}) = h\Delta T_e = \frac{\dot{m}_b h_{fg}}{A} \quad (6)$$

This indicates the heat transfer rate from the heating element to the water is the same as the evaporative heat transfer rate [17].

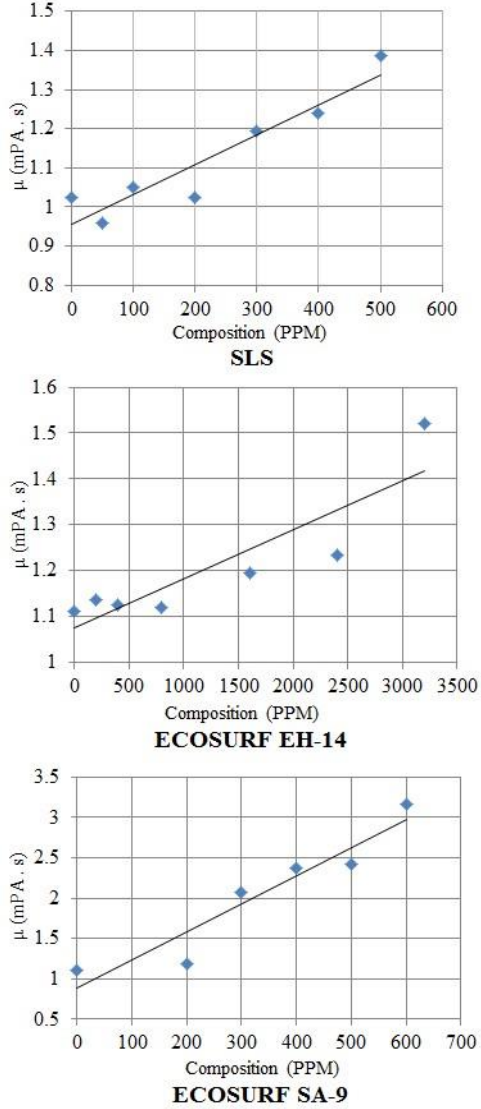


Fig. (2). Variation of viscosity with concentration for aqueous surfactant solutions [26, 27].

Eqn. 7 is developed by Rohsenow and is the first and most widely used correlation for nucleate boiling [17]. In this equation, surface tension and viscosity are main variables at surfactant tests.

$$q_s'' = \mu_l h_{fg} \left[\frac{g(\rho_l - \rho_v)}{\sigma} \right]^{1/2} \left(\frac{c_{p,l} \Delta T_e}{C_{s,f} h_{fg} Pr_l^n} \right)^3 \quad (7)$$

If one dimensional steady heat flow from the heater to the water is assumed, the heat flux through borosilicate beaker is given by Fourier's law [17]:

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$$q_s'' = -k_{borosilicate} \frac{\partial T}{\partial x} = -k \frac{\Delta T}{\Delta x} \quad (8)$$

With temperature difference between beaker bottom and beaker inlet is measured, the heat flux is calculated. This is the heat flux delivered to the liquid. Consistent values are obtained for q'' at Eqn. 5 and 6 and 8.

5. RESULTS

Fig. (2) shows the variation of viscosities with concentration for aqueous SLS, EH-14 and SA-9 solutions. An increase in viscosity with the increasing concentration is observed.

Fig. (3) shows the boiling observations. EH-14 and SA-9 looks more turbid during boiling compared to water and SLS. Fig. (4) shows the boiling curves of aqueous solutions of surfactants. Boiling curves are shifted to the left with the surfactant additions. An increase in concentration did not shift the boiling curve further after some concentrations.

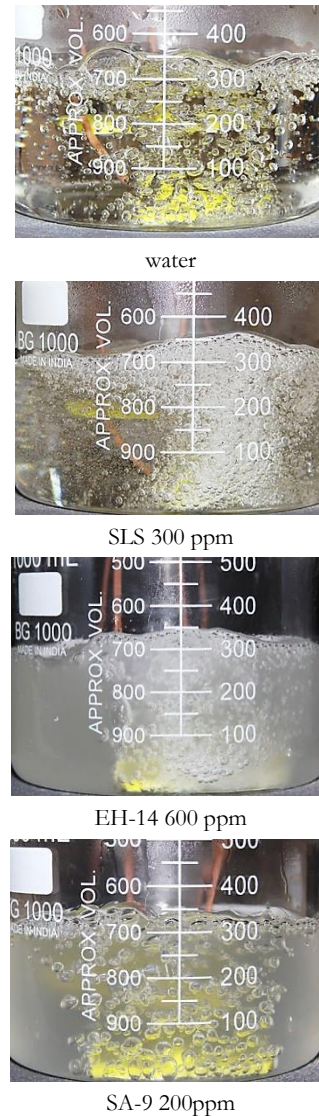


Fig. (3). Boiling observations at heat flux of 20 (kW/m²) [27].

6. DISCUSSION

In the previous studies of Dikici *et al.* [18-19] same surfactants were studied. However, in these studies, the boiling curves were drawn for selected concentrations which performed fastest depending on time required to reach to the boiling point. In this study, boiling curves of wider composition of surfactant solutions are drawn separately and they are compared to each other and boiling of water. The results of this study agreed that boiling curve of the surfactant solution depends on its concentration and every surfactant solution at given concentration behaves itself as new liquid having the common boiling curve at various level of subcooling [20].

6.1. Heat Transfer Coefficient, h

Fig. (2) shows the boiling curve of aqueous solutions of SLS, EH-14 and SA-9. 31%, 18%, and 10% lower wall superheats are found compared to water (for 400 ppm SLS, for 800 ppm EH-14 and for 200ppm SA-9 respectively). Boiling heat transfer coefficient (h) enhancements compared to water are 46%, 30%, and 21%. (for 400 ppm SLS, for 800 ppm EH-14 and for 200ppm SA-9 respectively). Increasing the concentration after 400ppm for SLS, 800ppm for EH-14 and 200ppm for SA-9 has no further improvement.

The lowered surface tension with surfactants reduces the nucleation radius, thus proceeding more active nucleation sites. It can also allow the departures of smaller sized bubbles [21].

Boiling observations showed that boiling with surfactant solutions compared to pure water is stronger. Bubbles in boiling surfactant solutions are smaller in size, activate continuously, and collapse rapidly.

6.2. Viscosity, μ

Viscosity increased with surfactant concentration for all compositions of (SLS, EH-14, SA-9). The obtained viscosity of 400 ppm SLS, 800 ppm EH-14, and 200 ppm SA-9 concentrations are 1.24 mPa·s, 1.04 mPa·s and 1.1 mPa·s respectively. These values can be considered close because temperature has a big influence on viscosity. Water viscosity at 20°C is 1.002 mPa·s and reduces 23% in only 10°C temperature difference.

SLS and EH-14 are soluble in water whereas SA-9 is dispersible in water. It is harder to blend the mixture thoroughly with magnetic stirrer when mixture is not water soluble. This might be the reason of the higher viscosity values for SA-9 at higher concentrations. Surfactant solution's Newtonian or Non-Newtonian behavior also effects when the viscosity values measured at increased concentrations. If the solution viscosity does not change considerably at higher concentrations, that means solution reveals the Newtonian fluidic behavior [1]. Viscosity of Non-Newtonian surfactant solutions are increased with the surfactant concentrations [1]. Several studies have also concluded that surfactants with pre-micellar or dilute concentrations cause no significant change in the dynamic viscosity of solution [1, 22, 23] whereas viscosity

reduces heat transfer in viscous solutions showing the non-Newtonian fluidic behavior.

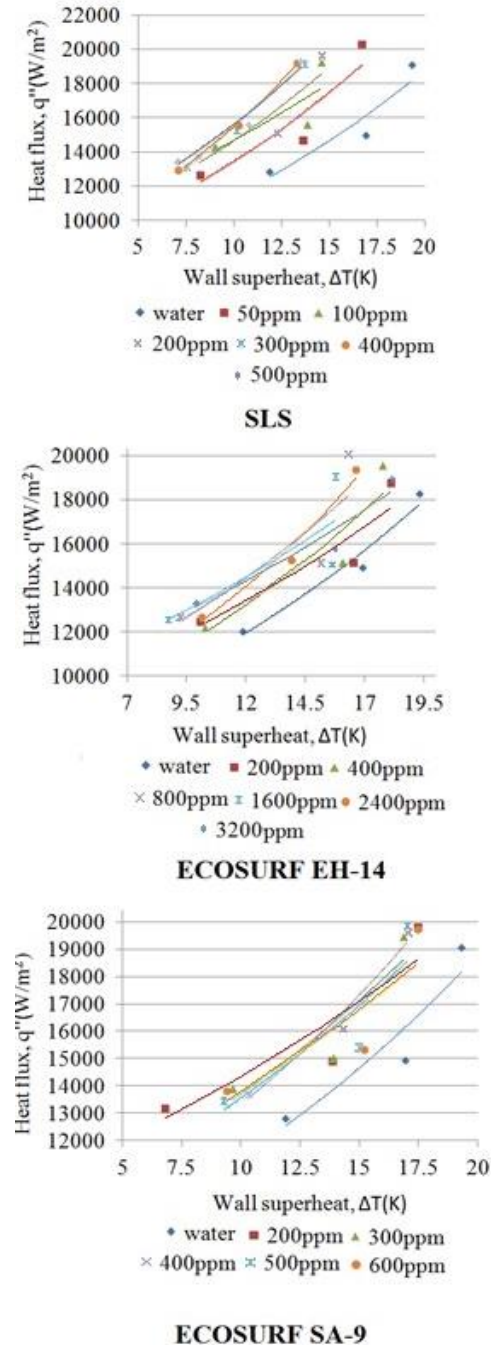


Fig. (4). Boiling curves of aqueous solutions of surfactants [27].

The effect of both the surface tension and the kinematic viscosity of surfactant mixture can explain the features of heat transfer at boiling of surfactant solutions. Hestroni *et al.* [12] worked with cationic surfactant Habon-G and showed that further increase in additive concentration leads to an increase in the viscosity and a decrease in the heat transfer coefficient. Therefore, the enhancement of heat transfer is connected to the decrease in surface tension values at low surfactant concentration, whereas the decrease in heat transfer at higher

surfactant concentrations is related to the increase in viscous characteristics [12].

6.3. Molecular Weight

According to Henneberg *et al.* [24], the number of active nucleation sites may be dominated by diffusion of surfactant molecules. It was found that surfactants with lower molecular weight diffuse faster than those with higher molecular weight [24]. It can be seen that the SLS has the lowest molecular weight among the surfactants, and this argument agrees well with diffusion controlled mechanism. However, for the nonionic surfactants, contradiction is observed.

6.4. Cloud Point

Fig. (4) shows boiling observations of water and the surfactant solutions. EH-14 and SA-9 (Nonionic surfactant solutions) are more turbid compared to water or anionic SLS. Cloud point is the temperature above which an aqueous solution of a water-soluble surfactant becomes turbid [25]. Cloud points are characteristic of nonionic surfactants. Anionic surfactants are more water-soluble than nonionic surfactants and will typically have much higher cloud points (above 100°C). Wetting, cleaning and foaming characteristics of a surfactant solution can be different above and below the cloud point. In general, nonionic surfactants show optimal effectiveness when used near or below their cloud point [25]. Low-foam surfactants should be used at temperatures slightly above their cloud point [25]. Therefore, cloud point can affect the boiling characteristics as well.

7. CONCLUSIONS & FUTURE WORK

Nucleate pool boiling of surfactant solutions is investigated. It is found that the boiling heat transfer enhancement of SLS is higher than that of EH-14 and SA-9 compared to water. The heat flux, surfactant concentration, surface tension, and molecular weight are considered to be the main factors that lead to enhancement in nucleate pool boiling.

Further experimentation with different binary combinations are recommended along with accurate measurements of dynamic surface tension, density and kinematic viscosity of surfactant solutions. Besides the effects of dynamic surface tension, concentration of surfactant, CMC, its chemistry (anionic nature for SLS and nonionic for EH-14 and SA-9), surface wetting, Marangoni convection, surfactant adsorption and desorption and foaming must be considered to have significant influence on boiling [18, 19]. Also, testing at higher heat fluxes will demonstrate the effect of surfactant at DNB. It is important to note that, the presence of a dissolved surfactant may cause issues in the various applications of boiling and condensation. These issues (foaming, accumulations, and effects on chemical treatment) should also be investigated.

Results prove that there is an important possibility to enhance the boiling application processes by environmentally friendly EH-14, and SA-9 additives. Experimentation can be extended

for searching other surfactants in order to find their most efficient quantity in water for boiling heat transfer.

ACKNOWLEDGEMENTS

All the work was conducted in the Clean Energy Laboratory at ERAU. We thank Remelisa Esteves and Nonso Onukwaba for viscosity experiments. The grant and support provided by ERAU are gratefully acknowledged. We acknowledge ASME for being the original publisher.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

NOMENCLATURE

A area, m²

c_p specific heat at constant pressure J/kg · K

C_{s-f} boiling constant corresponding to different surface–liquid combinations

CMC Critical micelle concentration, ppm

F force, N

g gravitational acceleration, m/s²

h convection heat transfer coefficient, W/m²K

h_{fg} latent heat of vaporization, J/kg

k thermal conductivity, W/m · K

m mass, kg

\dot{m} mass flow rate, kg/s

N Rate of formation of vapor nuclei

q heat transfer rate, W

q'' heat flux, W/m²

Pr Prandtl number

T temperature, °C

t time, s

x distance, m

GREEK LETTERS

ρ density (kg/m³)

σ surface tension (N/m)

μ viscosity (kg/(s · m))

SUBSCRIPTS

b boiling

e excess

l liquid

s surface

sat saturation

sol solution

surf surfactant

v vapor

w water

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