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Effect of substrate temperature on evaporation of ethanol-water sessile droplet

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Abstract

The evaporation dynamics of an ethanol-water (E 50% + W 50%) binary sessile droplet of volume 5μl is studied experimentally and theoretically. The focus of this work is to investigate the effect of the substrate temperature (T_s) on the evaporation dynamics. A cellulose acetate tape with a surface roughness of 668 nm is used as the substrate. Two distinct evaporation stages are observed at T_s = 25 ^{0}C, namely an early pinned stage and a later receding stage. The higher volatile ethanol evaporates faster leading to a nonlinear trend in the evaporation process at the early stage. In contrast, at T_s = 60 ºC, three distinct phases are observed, which are the early spreading stage, an intermediate pinned stage and a late receding stage of evaporation. We have developed a theoretical model that combines evaporative diffusion, natural convection and passive transport of vapour, which predicts the experimental behaviour well.

Keywords: Binary sessile drop, Evaporation, Spreading dynamics

1. Introduction

The investigation of the wetting and evaporation dynamics of sessile droplets has seen a lot of advancement in recent years. Several works have cited practical applications ranging from industrial to biological [1]. The research on pure fluids at various substrate temperatures has been carried out for years, and empirical models have been developed combining the diffusion and convective transport models which show good agreement with experimental studies [2,3,4]. However, the droplet dynamics are different for binary components [5]. While the droplet volume decreases monotonically in case of pure fluids, in a binary component, the evaporation occurs in three distinct stages. The first and last stages are dominated by the evaporation of highly volatile and less volatile components, respectively. At the intermediary stage, the volume remains constant with changing contact angle. In Refs. [6] and [7] water-ethanol droplets were studied on gold and poly-methyl-methacrylate (PMMA) substrates, respectively and similar behaviours were observed as in Ref. [5]. It was observed that the wetting hysteresis and initial evaporation of more volatile component have a significant impact on the change in evaporation mode. In Ref. [8], water-methanol droplets on a smooth polymer coated substrate were studied. The initial stage was dominated by the evaporation more volatile component (methanol), but it was also observed that a small amount of methanol remained, which influenced the wetting at later stages. They observed four distinct stages in the contact angle dynamics and the spreading characteristics of the droplet.

In all the studies, the substrate temperature was maintained at the room temperatures. The substrate properties like surface roughness were also not reported which have an impact on the wetting hysteresis and can change the evaporation behavior. Recently, we investigated the evaporation of a sessile droplet of ethanol-water binary mixture by varying its composition and substrate temperature [9]. In the present study, we used a fixed concentration ethanol-water (E 50% + W 50%) binary sessile droplet to study the effect of substrate temperature on the evaporation dynamics. A cellulose acetate tape with a surface roughness of 668 nm at 25 ^{0}C and 641 nm at 60 ^{0}C is used, thereby maintain uniformity even at higher temperatures. Subsequently, the theoretical model has been developed explaining the observed evaporation rate.

2. Experimental set-up

The experiment involves the investigation of an evaporating water-ethanol sessile binary drop from a heated substrate. The substrate is temperature controlled and connected to PID regulated heaters. The setup is inside a cell to avoid any disturbances from the external air flow. Evaporation takes place at one-atmosphere pressure and at an ambient temperature of 25 ^{0}C with a relative humidity of 36% monitored by an HTC 288-ATH hygrometer. The customised experimental set-up is provided by Holmarc. The schematic diagram of the setup is shown in Figure 1.

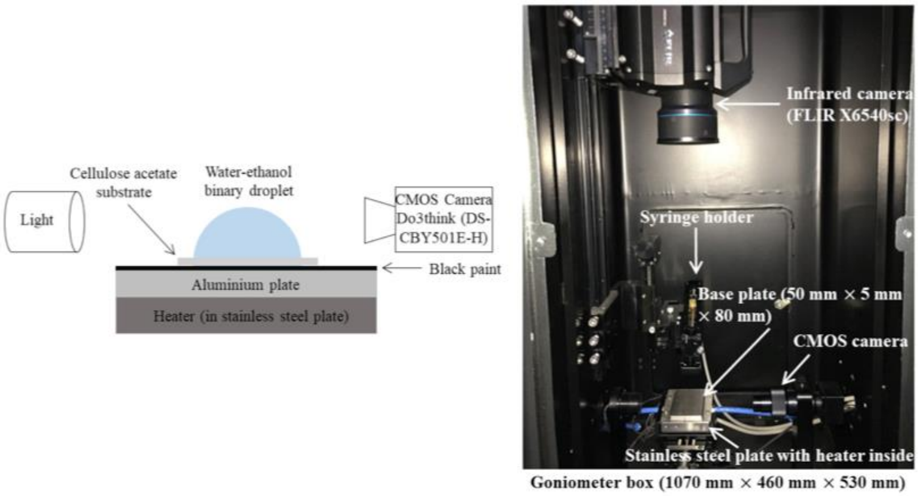

Figure 1: Schematic diagram of the experimental setup (left) and the actual image of the goniometer (right).

The binary liquid used is a water-ethanol mixture of various compositions by volume. The drop is placed on a multi-layered set-up which includes a stainless steel block (100 mm × 80 mm × 15 mm) at the base with two PID regulated electrical heaters inside. The substrate consists of an aluminium plate of thickness 5 mm with a black paint coating on which a cellulose acetate tape of thickness 0.063 mm (63 microns) is pasted and over which the drop is placed.

For the substrate, two materials were considered, a PTFE sheet of thickness 75 microns and the cellulose acetate tape of thickness 63 microns. The surface roughness of cellulose acetate is more uniform at higher temperatures. A complementary metal oxide semiconductor (CMOS) camera by Do3think (model number DS-CBY501E-H) is used to record the droplet evaporation. The videos are recorded at 10 frames per second (fps) with a spatial resolution of 1280 × 960 pixels.

The controller is adjusted to achieve the required temperature on the aluminum plate, and the measurement is made using an IR gun (+/- 0.5 ºC error). The heater is kept on for an hour before each experiment in order to achieve a steady state condition. The tape is then applied and kept for 10 minutes to attain thermal equilibrium with the substrate before the drop is placed. The small change in temperature at the surface of the tape is ignored owing to the small thickness of the tape. The droplet volume is kept constant at 5 µl for all the experiments. A motorised pump controls the volume to be pumped (error < 1 %) at the desired flow rate. The syringe (needle outer diameter = 1.59 mm) is cleaned with acetone and allowed to dry before performing each experiment. The experiments are repeated six times for each test conditions by changing the tape every time and placing the drop at different locations.

Image processing is done using MATLAB to extract the droplet boundary at different time intervals. The in-house code allows the calculation of the dynamic contact angle, height and wetting radius of the evaporating drop with time. The volume at different times is calculated by assuming that the drop is of spherical-cap shape. The maximum error in the readings (contact angle, drop height and radius) is observed to be < 6%. The actual dimension of the drop is calculated by calibrating the camera, and the scaling factor is observed to be 227.27 pixels/mm.

3. Results

We investigated the evaporation dynamics of E 50% + W 50% binary droplet at different substrate temperatures T_S. The contours of the droplet at a different normalised time, t/t_e, show different spreading behaviour at $T_S = 25\ {}^0$C, $T_S = 40\ {}^0$C and $T_S = 60\ {}^0$C, which can be observed in Figure 2. Here, t_e is the total evaporation time of the droplet. It can be seen that negligible spreading is observed for $T_S = 25\ {}^0$C and 40 ^{0}C. For $T_S = 25\ {}^0$C, the droplet recedes at $t/t_e > 0.2$, prior to this it is pinned. For $T_S = 40\ {}^0$C, the droplet remains pinned at the later stage ($t/t_e > 0.2$), but the right contact angle (with respect to the view) recedes slightly, and the droplet becomes asymmetrical at the early stage ($t/t_e \leq 0.2$). In contrast, the spreading is more at $T_S = 60\ {}^0$C during $0 \leq t/t_e \leq 0.2$. but at the later stage ($t/t_e > 0.2$), the droplet is pinned during ($t/t_e = 0.2 - 0.6$) and then recedes at $t/t_e > 0.6$. From the experimental data, it is found that the lifetime of the droplet decreases from 1035 s to 38 s with the increase in the substrate temperature from $T_S = 25\ ^0$C to 60 ^{0}C as shown in Figure 3.

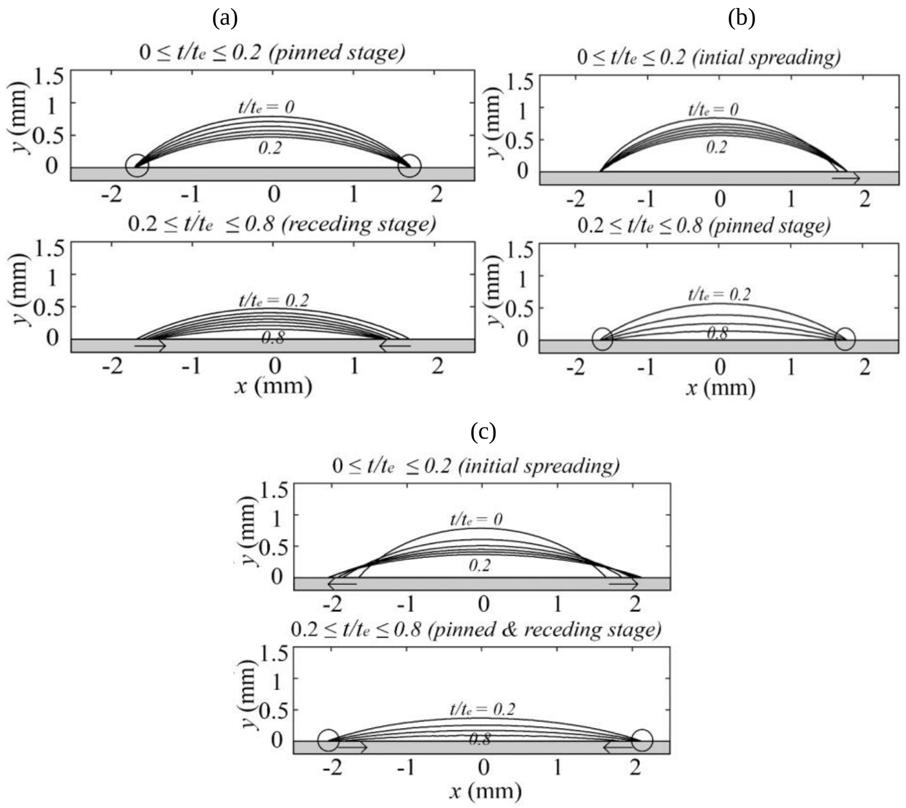

Figure 2: Comparison of the contours of a binary droplet of composition (E 50% + W 50%) at different substrate temperatures. (a) $T_S = 25\ ^oC$, (b) $T_S = 40\ ^oC$ and (c) $T_S = 60\ ^oC$.

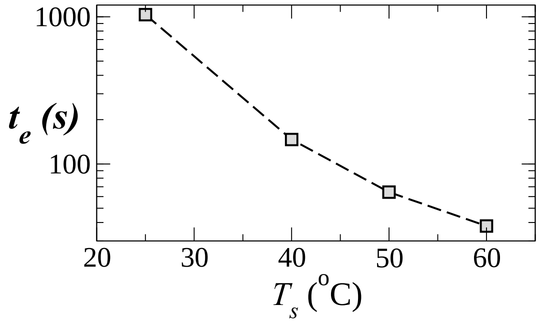

Figure 3: The variation of the total evaporation time, t_e (in seconds) of a droplet of (E 50% + W 50%) binary mixture with the temperature of the substrate, T_S.

The non-monotonic evaporation behaviour of a ethanol-water binary droplet of composition E 50% + W 50% at different substrate temperatures, $T_S = 25\ ^oC$, 40 ^{o}C and 60 ^{o}C is shown in the Figure 4. In that figure, the variation of the droplet height (*h* in mm), the wetting diameter (*D* in mm), and the droplet volume normalised with the initial volume of the droplet (V/V_0) are plotted against the normalised evaporation time (t/t_e). It can be seen that at $T_s = 25\ ^oC$, the droplet wetting diameter decreases during the entire evaporation process and there is no initial spreading, whereas at $T_s = 40\ ^oC$, the wetting diameter remains almost constant. In contrast, at $T_s = 60\ ^oC$, the droplet spreads faster at early times (D increases) and then it becomes constant. The presence of interface undulations increases the error in profile evaluation, and hence the assumption of spherical cap profile is not valid at the end stage of evaporation.

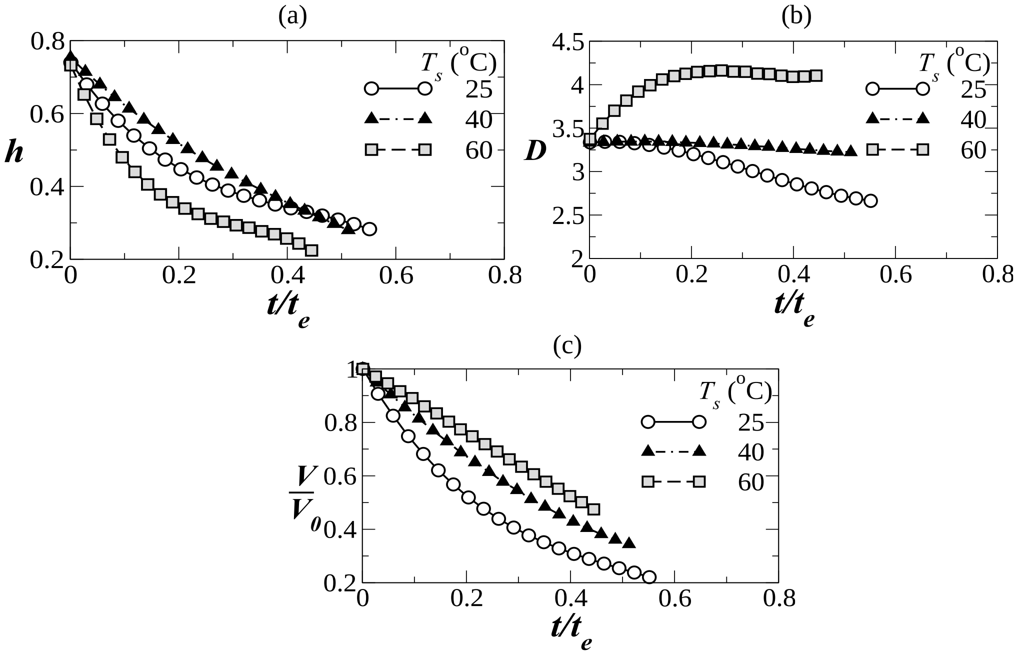

Figure 4: Variations of (a) the height (*h*) in mm, (b) the wetting diameter of the droplet (*D*) in mm, and (c) the normalised volume with initial volume of the droplet (*V/V_0*) versus time normalised with the lifetime of the droplet (*t_e*) at different substrate temperatures.

4. Theoretical modelling of droplet evaporation rates

We have developed theoretical evaporation models for sessile droplets for the binary mixture (E 50% + W 50%) and compared the theoretical predictions with our experimental results. The evaporation of binary ethanol-water droplets is dependent on three processes, namely the molecular diffusion, the convective mass transfer and the convective air flow induced passive vapour transport.

We assumed that all the droplets to have a spherical cap profile [8] during the evaporation process, which in turn implies that $\theta_l = \theta_r = \theta$. Thus the experimental droplet volume can be calculated as,

$$V(t) = \frac{\pi R^3 (1-\cos\theta)^2 (2+\cos\theta)}{3\sin^3\theta} \tag{1}$$

where, R is the wetting radius of the droplet and θ is average contact angle, which is found to be less than 90^0 in the present configuration. The rate of evaporation of the binary mixture due to diffusion is given by Refs. [3] and [4].

$$\left(\frac{dm}{dt}\right)_d = \pi RDM[C_{sat}(T_S) - C_\infty(T_\infty)] f(\theta) \tag{2}$$

where, *M* is the molecular weight of the mixture, *D* is the diffusivity at mean temperature $(Ts + T_\infty)/2$ and $f(\theta) = 1.3 + 0.267\theta^2$ for $\theta < 90^0$. Here, $C_{sat}(T_s)$ is vapour concentration at the liquid-vapour interface for the saturated condition, and $C_\infty(T_\infty)$ is the vapour concentration far away from the droplet.

The evaporation rate due to convection is given in Ref. [10] as

$$\left(\frac{dm}{dt}\right)_c = h_m A_S M[C_{sat}(T_S) - C_\infty(T_\infty)], \tag{3}$$

where, h_m is the convective mass transfer coefficient, A_s is the droplet surface area (a spherical cap profile is assumed). The convective mass transfer coefficient is calculated from the Sherwood number as

$$Sh_c \equiv h_m R / D \tag{4}$$

where D is the vapour diffusivity.

Thus the combined evaporation rate due to the diffusion and the convective from the droplet interface can be calculated as

$$\left(\frac{dm}{dt}\right)_d + \left(\frac{dm}{dt}\right)_c = h_{d+c} A_s M [C_{sat}(T_S) - C_\infty(T_\infty)] \tag{5}$$

where h_{d+c} is the combined diffusion and convective mass transfer coefficient, and can be evaluated using

$$h_{d+c} = \frac{Sh_{cor} D}{R} \tag{6}$$

The Sh_{cor} is obtained from the correlation developed by Kelly-Zion et al. [2]. As sessile droplet of a binary mixture is kept on a heated surface, free convection of air due to temperature gradient is considerable and the associated mass transfer flux can be expressed as

$$\left(\frac{dm}{dt}\right)_t = Y_v^s \left(\frac{dm}{dt}\right)_a \tag{7}$$

where Y_v^s represents the mass fraction of ethanol vapour above the free surface of the droplet and $(dm/dt)_a$ denotes the mass convection of air over the heated substrate not covered by the drop, which can be expressed as

$$\left(\frac{dm}{dt}\right)_a = h_m^a \pi R^2 \frac{M_a}{R_u} \left(\frac{p_\infty^a}{T_\infty} - \frac{p_s^a}{T_S}\right), \tag{8}$$

where air phase is approximated to be an ideal gas; M_a and h_m^a are the molecular weight and the mass transfer coefficient of air, respectively; R_u is the universal gas constant; p_∞^a and p_s^a are the partial pressures at the ambient and the plate surface, respectively.

The total evaporation rate for ethanol-water binary mixture sessile droplet on a heated substrate is the sum of the diffusion, convection, and passive transport and which is given by

$$\left(\frac{dm}{dt}\right) = \left(\frac{dm}{dt}\right)_d + \left(\frac{dm}{dt}\right)_c + \left(\frac{dm}{dt}\right)_t = h_{d+c} A_s M (C_{sat}(T_S) - C_\infty(T_\infty)) + Y_v^s \left(\frac{dm}{dt}\right)_a \cdot \tag{9}$$

The instantaneous mass evaporation rate of the individual component (ethanol and water) is calculated from the above relations, and binary VLE plots are used to calculate vapour pressure of solution and vapour phase mixture composition data. As the ethanol-water mixture is a non-ideal solution, the estimation of the excess molar volume is required, and it is expressed in terms of Redlich-Kister (R-K) correlations [11]. We obtain the volume of a droplet of the ethanol-water binary mixture at any instant as

$$V(t) = \frac{m_{droplet}(t)}{\rho_m(t)} \cdot \tag{10}$$

In Figure 5, we have plotted the experimental and theoretically obtained V/V_0 against t/t_e for the binary mixture (E 50% + W 50%) at T_s = 25 ºC, 40 ºC, and 60 ºC. Excellent agreement between experimental and theoretical results is obtained as evident in the Figure 5.

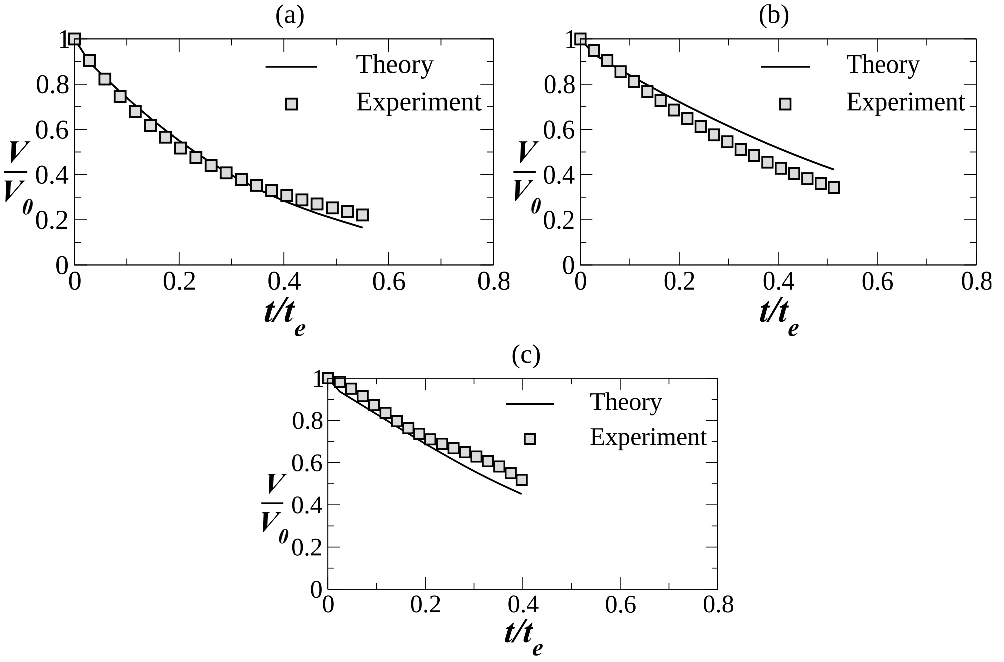

Figure 5: Comparison of the theoretical and experimental results for the binary droplet of (E 50% + W 50%) at (a) T_s = 25 ⁰C, (b) T_s = 40 ⁰C and (c) T_s = 60 ⁰C.

5. Conclusions

Experimental investigation of sessile droplets of a binary mixture (E 50% + W 50%) on a cellulose acetate tape is performed using a customised goniometer at different substrate temperatures: T_s = 25 ºC, 40 ºC, and 60 ºC. The higher volatile ethanol evaporates at the early stage leaving the less volatile water which evaporates at a relatively slower rate at the late stage, leading to a nonlinear trend in the evaporation process. Three distinct stages, an early spreading stage, an intermediate pinned stage, and a late receding stage are observed during the evaporation of a binary droplet. The evaporation dynamics of the binary drop at T_s = 25 ºC and 40 ºC are similar, whereas the behaviour is very different at a high substrate temperature (at T_s = 60 ºC).

References

[1] Brutin, D. & Starov, V. 2018 Recent advances in droplet wetting and evaporation. *Chem. Soc. Rev.* **47** (2), 558-585.

[2] Kelly-Zion, P. L., Pursell, C. J., Vaidya, S. & Batra, J. 2011 Evaporation of sessile drops under combined diffusion and natural convection. *Colloids Surf.* A. **381** (1-3), 31-36.

[3] Sobac, B. & Brutin, D. 2012 Thermal effects of the substrate on water droplet evaporation. *Phys. Rev. E.* **86** (2), 021602.

[4] Carle, F., Semenov, S., Medale, M. & Brutin, D. 2016 Contribution of convective transport to evaporation of sessile droplets: empirical model. *Int J. Therm. Sci.* **101**, 35-47.

[5] Sefiane, K., Tadrist, L. & Douglas, M. 2003 Experimental study of evaporating water-ethanol mixture sessile drop: influence of concentration. *Int. J. Heat Mass Transf.* **46** (23), 4527-4534.

[6] Cheng, A. K. H., Soolaman, D. M. & Yu, H-Z 2006 Evaporation of microdroplets of ethanol-water mixtures on gold surfaces modifed with self-assembled monolayers. *J. Phys. Chem. B.* **110** (23), 11267-11271.

[7] Wang, Z., Peng, X-F., Mujumdar, A. S., Su, Ay & Lee, D-J 2008 Evaporation of ethanol-water mixture drop on horizontal substrate. *Dry. Technol.* **26** (6), 806-810.

[8] Brutin, D. 2015 *Droplet wetting and evaporation: from pure to complex fluids.* Academic Press.

[9] Gurrala, P., Katre, P., Balusamy, S., Banerjee, S. & Sahu, K. C. Evaporation of ethanol-water sessile droplet of different compositions at an elevated substrate temperature. International Journal of Heat and Mass Transfer 145, 118770.

[10] Kelly-Zion, P. L., Pursell, C. J., Wassom, G. N., Mandelkorn, B. V. & Nkinthorn, C. 2018 Correlation for sessile drop evaporation over a wide range of drop volatilities, ambient gases and pressures. *Int. J. Heat Mass Transf.* **118**, 355-367.

[11] Jimenez, J., Manrique, J. & Martinez, F. 2004 Effect of temperature on some volumetric properties for ethanol + water mixtures. *Rev. Colomb. Cienc. Quim. Farm.* **33** (2), 145-155.