Influence of surface properties on the dynamic behavior of impacting droplets

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An analysis is performed here of the mechanisms governing the coupling between surface and wetting dynamics of spreading droplets on rough surfaces which may give considerable insight into the effects of wettability. This study is part of a research aimed at characterizing the influence of the nature of the target surface on impacting droplets, as an attempt to determine which a priori known properties of the system liquid-target-vapour can be used to predict the spread behavior. Measurements of the equilibrium contact angle showed that the wettability is strongly related with the surface roughness, although this relation differs for small and strong roughness. Also, it is shown that wettability influences the spreading mechanism, mainly for the last stages and for low impact velocities.

1. Introduction

The dynamic behaviour of an individual droplet impacting onto a solid surface includes several individual phenomena, such as deformation, fingering, splashing and rebound. The onset and physical description of those phenomena are usually characterized based on dynamic similarity arguments making use of dimensionless numbers characterizing the relative magnitude of the forces acting upon the surface of the droplet (Reynolds, Weber and Ohnesorge numbers), *e, g.,* Chandra and Avedisian [1], Bussman *et al.*[2], Marmanis and Thoroddsen [3]. Estimative of these numbers are, in turn, obtained by scaling the forces (surface tension, shear and gravity) with physical properties of the liquid (density, viscosity and surface tension) and considering all lengths and velocities proportional to the diameter and velocity at the instant of impact, respectively. However, the physical properties of the impacting surface such as temperature, roughness or inclination alter the boundaries of the problem and similarity arguments cannot be applied. One of the most interesting features observed in the spreading of non-volatile droplets on solid target is the effect of the nature of the surface on the wetting dynamics.

Recent investigations on droplet impact report on the influence of the target surface. Wu [4], Stow and Hadfield [5] and Mundo, Sommerfeld and Tropea [6] obtained correlations to predict the onset of splash, based on a splash parameter, which depends on the mean roughness, R_a. Range and Feuillebois [7] showed that the onset of splashing is significantly altered by other characteristics of the surface, such as the surface material and profile, which are not included in R_a. The wettability, measured by the contact angle of the liquid film with the surface, may be a key parameter in this analysis. Kandlikar and Steinke [8] investigated the influence of temperature, roughness and material of the target surface on the dynamic advancing and receding contact angles; Rioboo, Marengo and Tropea [9] studied the influence of surface wettability on droplet spreading. However, it appears from the reviewed literature, that the effect of the nature of the target surface on the dynamic behaviour of droplets impacting onto a surface is not fully understood and more fundamental work is still necessary.

This paper is part of a research study aimed at to account for the complexity introduced by the influence of non-scaled parameters on the description of droplet impact. In this context an experimental installation was built as described by Moita and Moreira [10] where droplets are generated at the tip of a hypodermic needle and fall by gravity onto a cold and flat surface. The time history of droplet deformation is recorded by a high-speed camera triggered by the passage of the droplet through a laser beam hitting onto a photodiode. This paper follows previous experiments reported in Moita and Moreira [10, 11] which considered deformation and splash of water and Diesel droplets impinging onto dry flat surfaces of different materials and with different surface profiles.

2. The relation between wettability and the nature of the surface

When a droplet impacts onto a solid surface, a liquid film is formed which spreads out with a retarding velocity, due to the influence of both, surface tension and surface forces. As a result of those forces, the liquid film attains a maximum spread when it comes to a stop. At that instant, the liquid mass accumulates around the splat periphery, giving rise to a pressure force, which drives recoiling. The film follows, then, a periodic motion until it comes to equilibrium when all the impact energy is dissipated. At equilibrium, only surface forces act upon the system formed by the liquid, the solid surface and the surrounding vapour. Young law provides a relation between those forces:

$$\sigma_{lv}.\cos\theta + \sigma_{ls} = \sigma_{sv}$$

where θ is the angle of contact that the liquid makes on the solid, σ_{lv}, σ_{ls}, and σ_{sv} represent the interfacial tensions at the boundaries between liquid (l), solid (s), and vapour (v). The contact angle, θ, is usually referred as the *wettability* of the surface. The liquid is seemed non-wetting when $90^o < \theta < 180^o$ and wetting, when $0^o < \theta < 90^o$ (see Figure 1). $\theta = 0^o$ and $\theta = 180^o$ correspond to complete wetting and non-wetting, respectively.

Fig. 1 Liquid droplet in equilibrium: definition of the contact angle.

Young's equation shows that the interfacial tensions are the solely parameters governing *wettability*. However, it has been shown that, despite the equation gives a practical insight into the phenomena, external fields may be present which alters the contact angle. The effects of the nature of the surface on spreading are interpreted here as indirect results of the effects upon the variable *wettability*, *e. g.*, Adão *et al*., [12].

The effects of gravity and surface deformation (*e. g.*, Sasges and Wald [13], Wald and Sasges [14], Sakai and Fujii [15]) are expected to be negligible in our study, since we are considering horizontal surfaces, low impact energies, negligible interfacial chemical reactions and diffusion processes are unlikely to occur. Chemical inhomogeneities due to consistency and cleanness of the surface, were minimized by carefully preparing and always characterising the target area before each experiment. Finally, another effect on Young's equation is due to surface roughness. Young's equation applies to ideal surfaces that are perfectly smooth. The contact angle measured on a rough surface (called the Wenzel [16] angle, θ_w) does not obey Young's equation, although it may be related to the equilibrium (Young) angle θ_y:

$$\cos\theta_w = r \cos\theta_y$$

where r is the ratio of the true wetted area to the apparent area. Wenzel model does not consider the contact angle histeresis caused by roughness. Histeresis is defined as the difference between the advancing and receding angles and arises because the liquid-vapour interface does not retrace its original path when recedes on the solid, so that the spreading is thermodynamically irreversible, *e. g.,* Zou and Hosson [17]. The analysis presented here is an attempt to consider the equilibrium angle, since that is the property of the pair liquid-target material which can be known a priori.

Contact angles at equilibrium were measured for several rough surfaces and liquids. The measurements were made with sessile drops inside a thermostatted ambient chamber (Ramé-Hart Inc., USA, model 100-07-00), with quartz windows, to avoid optical distortion, previously saturated with the liquid to be studied (water and diesel oil) at a temperature of 20 ±1ºC. After the drop was deposited at the surface, its image is recorded using a colour video camera (JVC Colour TK-1070) mounted on a Wild M3Z microscope, which allows a magnification of 40 times. The video signal was transmitted to an image processor - Video Pix Framegrabber (Sun Microsystems) – that performs its digitalization in 640 x 480 pixels images, in a 256 grey level scale. The image acquisition and analysis was performed by a Sun Sparc station IPC, using the Axisymmetric Drop Shape Analysis software (ADSA for SunOS 1.0 Applied Surface Thermodynamics Research Associates, Toronto, Canada). According to Cheng, [18] the precision of the algorithm used by ADSA to obtain the contact angle is about ±0.1º. The variation of the contact angle with time was recorded for time intervals of six hundred seconds. The first image was taken about one second after deposition. For the first 100s, when the changes with time are more evident, the images were taken every ten seconds and the subsequent images were taken every twenty seconds. Time between images was about 0.72s. At least 8 measurements were taken for each pair liquid-impact surface in order to obtain average values. Furthermore the evolution of the average contact angles with time was obtained for each pair liquid-surface, by curve fitting and the final values were determined by extrapolation. Since the outcomes from droplets impact occur in a much smaller time scale (ms) it was considered that the final value for the contact angle was obtained by fitting the values obtained in the latest period of the measurement and extrapolating them for t = 0s.

In this study we consider the use of several target plates, made of dissimilar materials with different surface properties. A number of rough plates were tested, from polished surfaces to rough surfaces with different roughness profiles, namely roughness wavelength and shape of the asperities. Surface roughness was characterized with a mechanical profile meter (perthometer C5D) that measures mean roughness up to 250μm. Numerous measurements were performed along orthogonal directions in order to form a grid of small target areas with well defined roughness characteristics. Inhomogenities of the targets, characterised by variations of the mean roughness, were found to be smaller than 10%. Table 1 summarizes the mean roughness of the target plates used in the present study.

Figure 2 depicts the contact angles measured for water drops impacting onto glass, Perspex and aluminum targets. The Figure clearly shows that the contact angle is a property of the pair liquid-surface, which varies either with the material of the surface and/or with its roughness, as also observed by Kandlicar and Steinke [8] with water droplets impinging onto cooper and stainless steel surfaces. In general, the results show that the contact angle increases with mean roughness, which is to be contrasted with the observations of [8] who report a decrease of the contact angle. An exception occurs for the rougher glass surface, which may be due to the comparatively rougher texture of the target used in the present work as reported by Hitchcock *et al.* [19], despite the authors observed a linearly increase of wetting with the texture parameter, Ra/λ, where λ is the average wavelength of the surface textures.

Table 1 Target surfaces used in this study

Surface		***Ra [μm ±10%]***
Glass	Smooth	≈ 0
	Random profile	2.01
		2.7
Perspex	Smooth	≈ 0
	Random Profile	1.0
		1.7
		1.9
Aluminum	Random Profile	1.52
		3.0
	Regular Profile	2.22

Furthermore, these results support the analysis performed in previous work [11] and by other authors, that the mean roughness is insufficient to characterize the surface topography. However, for commercial surfaces like those used in this study, it is not trivial to define a characteristic wavelength because the roughness profile is, in general, very irregular.

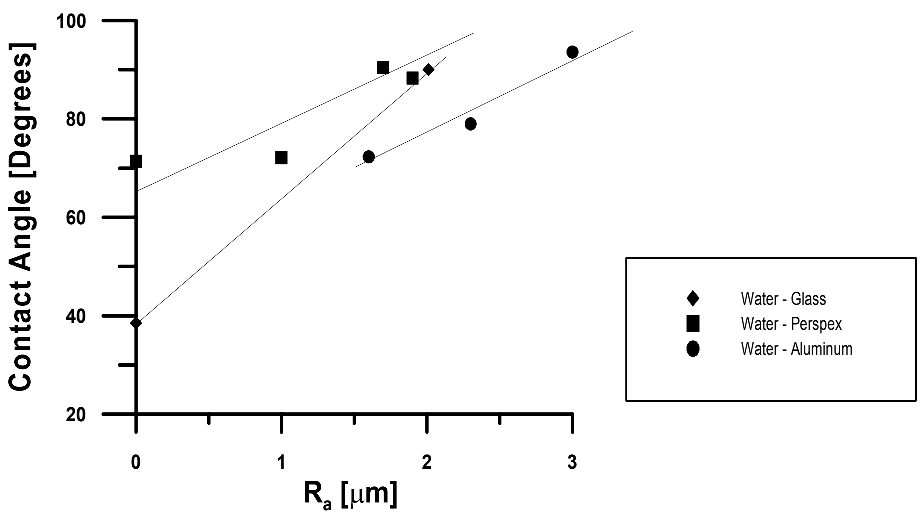


Fig. 2 Contact Angles for water onto different target surfaces.

3. The effect of wettability on spreading dynamics

Accordingly to Young's law, the *wettability* is an equilibrium thermodynamic variable that depends only on the interfacial tensions of the surfaces. However, despite the equilibrium is attained in a very short time (of the order of 10^{-4} to 10^{-1} sec), the contact angle is a dynamic variable and its variation determines the spreading behaviour, *e. g.* Dussan [20]. It has been suggested that the dynamic contact angle depends on the rate of spreading. Several semi-empirical and theoretical relations have been proposed in the literature, but all have the form of a power law $d(t)^{\alpha} \sim t$, where *d(t)* is the instantaneous diameter of the spreading droplet and *t* is the time, *e. g.*, Asthana and Sobczak [21].

The measured spreading data are plotted in Figure 3 as *ln d(t)* versus *ln t*. The plot shows that the experimental results do not follow the straight line consistent with the referred classical models, but rather exhibit a more complex spreading behaviour for the early times of spread, which is attributed to the effect of the nature of the target surface.

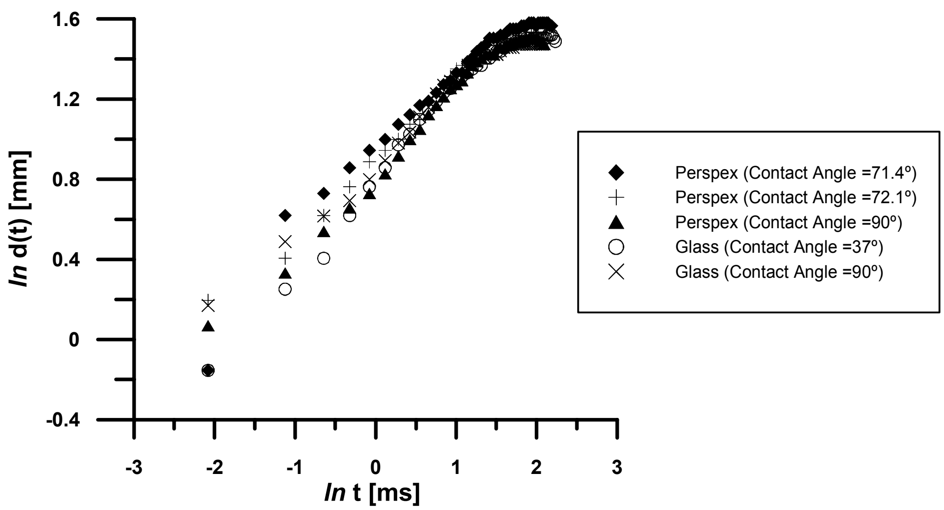
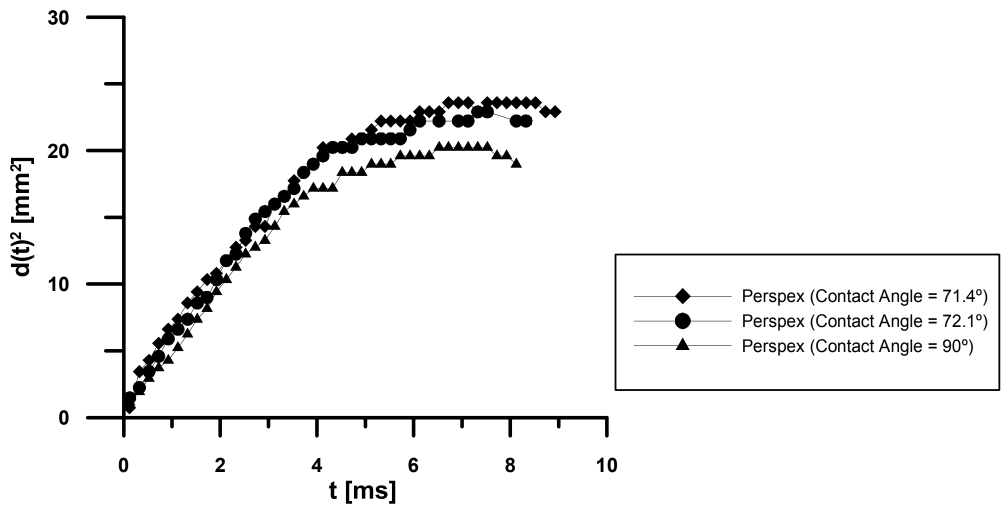


Fig.3 *ln d(t)* versus *ln t* for the measured spreading data.

It is worth mentioning that the spreading kinetics in a given system is strongly affected by the experimental conditions, and very different spreading kinetics can be measured for the same system depending upon the experimental technique (oxygen partial pressure, temperature, alloying technique, surface preparation, etc.). Thus, while the results in Figure 3 illustrate the complexity of the spreading phenomenon, they shall be analysed with care if the underlying mechanisms are to be explained. For example, the contact angles were also measured for Diesel oil droplets but it was observed a complete wetting off all the surfaces (contact angle near zero). This may be due to the presence of a precursor film on the target surface allowing faster spreading rates because it relaxes the no-slip boundary condition between the liquid and the surface *e. g.*, Abraham *et al.* [22] and Troian [23]. The phenomenon induces a super spreading behaviour and precludes the characterization of the system liquid-surface by the equilibrium contact angle, as also reported by Cohen-Stuart *et. al.* [24] and Randal [25].

Figure 4 shows the rate of area increase of the spreading film on perspex and glass targets, as the variation of $d^2(t)$ with time. The results show that the area increases linearly with time up to a plateau value, which changes with surface wettability, as in Zhou *et al.* [17]. In general the results in the Figure show that the rate of spreading increases as the contact angle decreases, as well as the plateau.

These results may be contrasted with those reported by Richard [26] and Mao *et al.* [27], who did not observed any difference of the maximum spreading diameter while changing the static contact angles. On the other hand, Fukai *et al.* [28] showed that the effect of wettability is important throughout all the spreading phase. Rioboo, Marengo and Tropea [9] also observed differences in the final stages of spreading, for non-wettable surfaces.



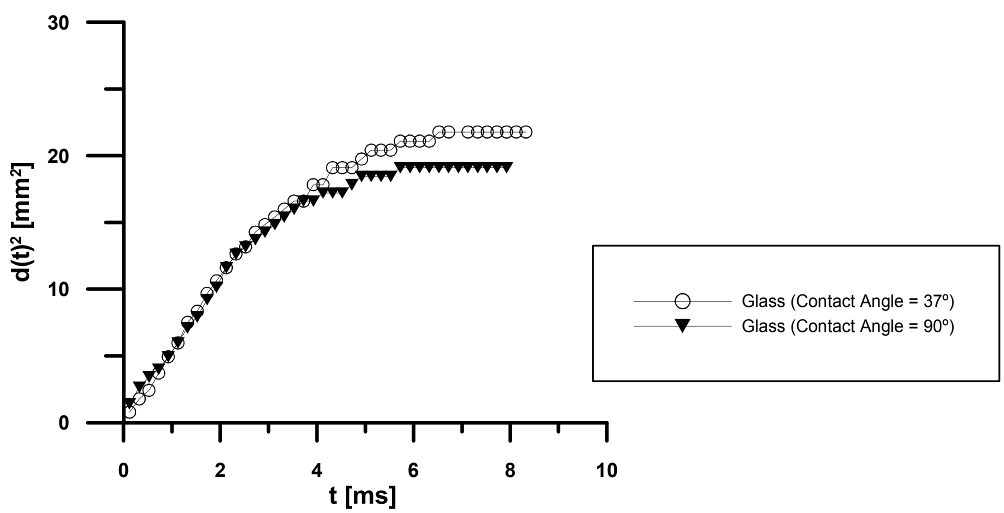


Fig.4 Spreading rate of water droplets with initial diameter of 3.2.mm impacting onto different surfaces with a velocity of 0.44 m/s.

However, it is worth mentioning that the experiments reported here consider very low impact velocities, for which the effects of wettability may be more noticeable. According to Pasandideh-Fard *et al.* [29], who established by mass and energy conservation, an equation to predict the maximum spreading rate involving the contact angle, stated that the wettability may eventually become negligible for high Webber numbers, but may be significant for low impact velocities. Figure 5 also shows that the maximum diameter of spread decreases monotonically with the wettability.

Therefore, although the wettability may not have negligible effects in some cases, the contact angles seem to play an important role in the spreading mechanism, which must be considered in modelling. Nevertheless, this is a highly complex process depending on a numerous factors and a universal model seems quite difficult to obtain.

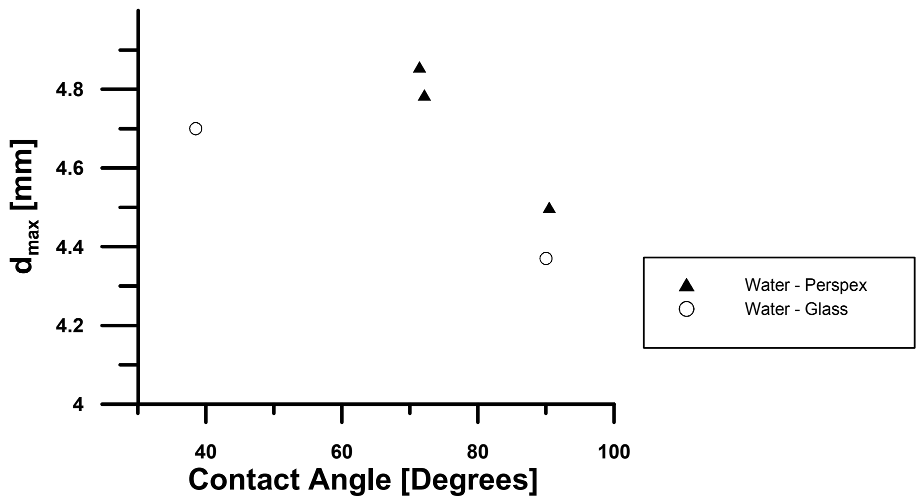


Figure 5 – Maximum spread diameter as a function of contact angles.

4. Summary

This paper is part of a research program aimed at studying the effects of the nature of the surface on the wetting dynamics of droplets impacting onto flat targets. The analysis presented here is an attempt to find the relevant properties of the thermodynamic system liquid-surface-vapour, which may be used if spreading dynamics is to be accurately predicted. Several targets made of different materials were used upon which droplets impact and the time variation of spreading was recorded with a CCD camera. The surface of the targets is characterized by its topography making use of a profile meter and by its wettability defined by the equilibrium contact angle as described by Young's law.

The results show that the wettability strongly depends on the mean surface roughness, and may be used as a characteristic parameter of the system, providing that a precursor film is not formed at the target surface during spread. For the low impact velocities considered here, it is suggested that the equilibrium contact angle describes the effects of surface roughness on the wetting dynamics. However, it is not still possible to find universal laws and other parameters shall further be considered, which account for the effects of surface irregularities of commercially available materials.

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References

[1] Chandra, S., Avedisian, C. T., 1991,.*Proc. R. Soc. Lond*., Ser. **A 432**, 13-41
[2] Bussman, M., Chandra, S., Mostaghimi, 2000, J., *Phys. Fluids*, vol.9, **12**, 3121-3132
[3] Marmanis, H., Thoroddsen, S. T., *1996*, *Phys. Fluids*, vo.8, **6**, 1344-1346
[4] Wu, Z. N., 1992, Ph.D. Thesis, Université Pierre et Marie Currie, Paris, France
[5] Stow, C. D., Hadfield, M. G., 1981, *Prc. R. Soc. of London*, Ser. **A 373**
[6] Mundo, CHR, Sommerfeld, M., Tropea, C., 1995, *Int. J. M. Flow*, vol.21, **2**, 151-173
[7] Range, Kai, Feuillebois, François, 1998, *J. Col. Int. Sci*, **203**, 16-30
[8] Kandlikar, Satish G., Steinke, Mark E., 2001, J. Ch. Eng. Res. Des., **74**
[9] Riboo, R., Marengo. M., Tropea, C., 2002, *Exp Fluids*, published online
[10] Moita, A. S. and Moreira, A. L. N., 2002, *SAE Technical Paper 2002-01-274.*
[11] Moita, A. S. and Moreira, A. L. N., 2002, *18th Annual Conference on Liquid Atomization and Spray Systems*.
[12] Adão, M. H., Saramago, B., Fernandes, A. C., 1999, *J. Col Int. Sci.*, **217,** 94-106
[13] Sasges, M., Wared, 1998, C. A., *J. Chem. Phys*., **109**, 3661-3670
[14] Ward, C. A. and Sasges, M. R., 1998, *J. Chemical Physics*, 109, 3651 – 3660, 1998.
[15] Sakai, H., Fujii, T., 1999, *J. Col. Int. Sci.*, **210**, 152-156
[16] Wenzel, R. N., 1936, *Ind. Eng. Chem*., **28**, 988
[17] Zou, X. B., Hosson, J. Th. M., 1995, *J. Mat. Res*. 10, 1984-1992
[18] Cheng, P., 1990, PhD Thesis, University of Toronto, Canada
[19] Hitchcock, S. J., Carrol, N. T., Nicholas, M. G., 1981, *J. Mat. Sci.*, **16**, 714-732
[20] Dussan, V. E.B. "The moving contact line: the slip boundary condition", *J. Fluid Mechanics,* 77, 665 – 684, 1976.
[21] Asthana, R., Sobczak, N., 2000, *JOM*, published online.
[22] Abraham, D. B., Cuerno, R., Moro, Esteban, 2002, *Phys. Ver. Lett.*, 88, 101-206
[23] Troian, S. M., Herbolzheimer, E. and Safran, 1990, *Rev. Lett.* 65, 333-336 (1990).
[24] Cohen-Stuart, M., Elmendorp, J. J. and Troian, S. M., 1990, J. P. Hulin, A. M. Cazabat, E. Guyon and F. Carmona, Eds., North Holland, Amsterdam, p. 29-43.
[25] Randal, M. H., 1998, *Silicones in Coatings II Conference*, Florida, USA, March 24-26, 1998.
[26] Richard, D., 2000, PhD Thesis, University of Paris, France
[27] Mao, T., Khun, D. C. S., Tran, H., 1997, *AIChE J.*, 43, 2169-2179
[28] Fukai, J., Shiiba, Y., Yamamoto, T., Miyatake, O., Poulikakos, D., Megaridis, C. M., Zhao, Z., *1995*, *Phys. Fluids*, vol.7, **2**, 236-247
[29] Pasandideh-Fard, M., Quiao, Y. M., Chandra, S. Mostaghimi, J., 1996, *Phys. Fluids*, **8**, 650-659