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Trickle bed hydrodynamics for (non-)Newtonian foaming liquids in non-ambient conditions

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ABSTRACT

Hydrodynamic studies on trickle-bed reactors at non-ambient conditions overwhelmingly addressed coalescing systems despite numerous industrial applications concern the processing of foaming liquids for which engineering data are scantier. To fill this gap, the effects of temperature and moderate pressure (non-ambient conditions) are reported in this study on the shift of the transition from trickle-to-foaming-pulsing flow regimes, on the two-phase pressure drop, the liquid holdup, and the pulse frequency and velocity for Newtonian (air–cetyltrimethylammoniumbromide (CTAB)) foaming and non-Newtonian (air–0.25% CTAB–carboxymethylcellulose (CMC)) foaming systems. At constant superficial gas velocity, the trickle-to-foaming-pulsing flow transition boundary was observed at lower superficial liquid velocity in comparison to non-foaming systems. The transition boundary shifted towards higher gas and liquid superficial velocities with increasingly temperatures and pressures. The pulse frequency increased with temperature and/or pressure whereas the pulse velocity increased with temperature but it decreased with increasing pressure. Respective comparisons with the coalescing alter ego, namely, air–water and air–CMC/water systems, showed that Newtonian and non-Newtonian foaming systems behaved qualitatively similarly regarding the effects of temperature and pressure as the coalescing systems.

1. Introduction

Trickle-bed reactors (TBR), which consist of fixed beds fed cocurrently downwards with gas and liquid streams, host a diversity of gas–liquid–solid reactive systems. For instance, TBR has been ushering for decades the oil industry where it is the battle horse in its refining operations. Among the various processes relying on trickle beds, their hydrodynamics when foaming systems are involved remains a poorly explored subject even though foams emanate very often in the petroleum, pharmaceutical and food industries [1]. Especially in the petroleum industry, foams play an important role in productivity and petroleum recovery and processing [2]. Previous experimental work on foaming liquids highlighted the differences of TBR hydrodynamics in comparison to non-foaming systems [3–5].

Experimental studies on foaming in trickle beds were initiated by Larkins et al. [6] and Weekman and Myers [7]. Till the late 1990s, the experimental work on flow regimes, pressure drop and liquid holdup were performed for a variety of foaming gas–liquid systems [3,8–13] but were restricted mainly to ambient conditions. However, Wild et al. [14] showed that the hydrodynamics of foaming systems can be dramatically influenced by increased pressures such as skyrocketing amplitudes of pressure fluctuations after a flow pattern shift beyond the transition line between trickle and foaming-pulsing flow regimes. Some systematic studies were triggered on the effect of pressure on TBR hydrodynamics with weakly and strongly foaming liquids [4,5,15–17], such as the trickle-to-pulsing flow regime transition, the pressure drop and liquid holdup up to 2 MPa. To the best of the authors' knowledge, pulse velocity and frequency at non-ambient conditions are missing in the open literature. At much elevated pressures up to 8.1 MPa, Larachi et al. [18] reported pressure drop and liquid holdup data using as a foaming system nitrogen–1% (w/w) ethanol/water. It is worthy of notice that none of the published literature addressed the incidence of elevating temperature on the evolution of TBR hydrodynamics with foaming liquids.

This work therefore presents a systematic study on the effect of temperature on the hydrodynamics of TBR for Newtonian and non-Newtonian foaming systems. The influence of temperature on the trickle-to-foaming-pulsing flow transition boundary, the two-phase pressure drop, the liquid holdup, and the pulse velocity and frequency are reported for the first time. The two-phase pressure drop and liquid holdup at the trickle-to-foaming-pulsing

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Nomenclature

$\Delta P/H$	two-phase pressure drop (Pa/m)
f_p	pulse frequency (Hz)
P	pressure (Pa)
T	temperature (°C)
u	superficial velocity (m/s)
V_p	pulse velocity (m/s)
Greek letters	
ε_L	liquid holdup
σ	surface tension
Subscripts	
G	gas phase
L	liquid phase
p	pulse

transition are also analyzed at elevated temperature and moderate pressure.

2. Experimental setup

The experimental setup was discussed in detail elsewhere [19] and schematically represented in Fig. 1. The experiments were performed in a bed of 107 cm high and 4.8 cm ID packed with 3 mm glass beads. As foaming systems, the air–aqueous cetyltrimethylammoniumbromide (CTAB, 6.25 ppmw or 0.17×10^{-4} mol/L) and air–6.25 ppmw CTAB–0.25% (w/w) aqueous carboxymethylcellulose (CMC) solutions were prepared and their behavior was compared to the air–water and the air–0.25% CMC base case solutions, respectively. To prevent fading of foaminess during the high temperature tests, CTAB was chosen because it is a non-volatile surfactant in the studied temperature range. For the CTAB-containing Newtonian solutions, the CTAB critical micelle concentration increased from 0.95×10^{-3} to 2.35×10^{-3} mol/L when temperature increased from 25 and 90 °C. Correspondingly, the surfactant concentrations expressed as a percentage of critical micelle concentration varied from 1.8% to 0.7% [20]. Fractional surfactant concentrations between 0.6% and 1.8% CMC were already high enough to turn the systems into foaming (air–CTAB/water) and strongly foaming (air–CTAB–CMC/water) ones and to dramatically alter the reactor hydrodynamics with respect to the air–water and air–CMC/water base cases. The distinction between foaming and strongly foaming systems was judged on the basis of visual observation during preparation of the solutions. Production of much more foams was noticed for air–CTAB–CMC/water with respect to air–CTAB/water under equal fluid throughput conditions.

Similar to the preparation procedure in Aydin and Larachi [19], the CTAB–CMC solutions were prepared by dissolving first CTAB and then powdered CMC in water at ambient temperature. The pseudoplastic rheological behavior was well represented by an empirical power-law relation. The consistency index, k, and the power-law index, n, were fitted for each temperature after measuring the solution shear stress–shear rate response on an Advanced Rheometric Expansion System (ARES) rheometer in the 0–1000 s^{-1} shear rate ranges. Table 1 displays the physicochemical properties of 0.25% (w/w) aqueous CTAB–CMC solution from 25 to 90 °C. The viscosity and the surface tension are sensitive to temperature where the effective viscosity drops of CTAB–CMC and CMC are expressed with consistency index, k, and the power-law index, n. The properties of the 6.25 ppmw CTAB solution are also given in Table 1 along with the aqueous CMC solution.

For the elevated temperature measurements, the liquid, prior to be routed to the reactor via a calibrated flowmeter was heated in a reservoir through a liquid preheater. The gas was supplied from a compressed air line up to a maximum pressure of 0.7 MPa. After passing through a preheater, the gas phase encountered the heated liquid phase at the top of the reactor. Both phases were introduced co-currently downwards through a distributor, which was designed to obtain a uniform distribution. At the reactor outlet both phases were intercepted in a separator where the gas phase was vented to the atmosphere via a calibrated flowmeter and the liquid phase was drained. Measurements were taken only when the desired steady-

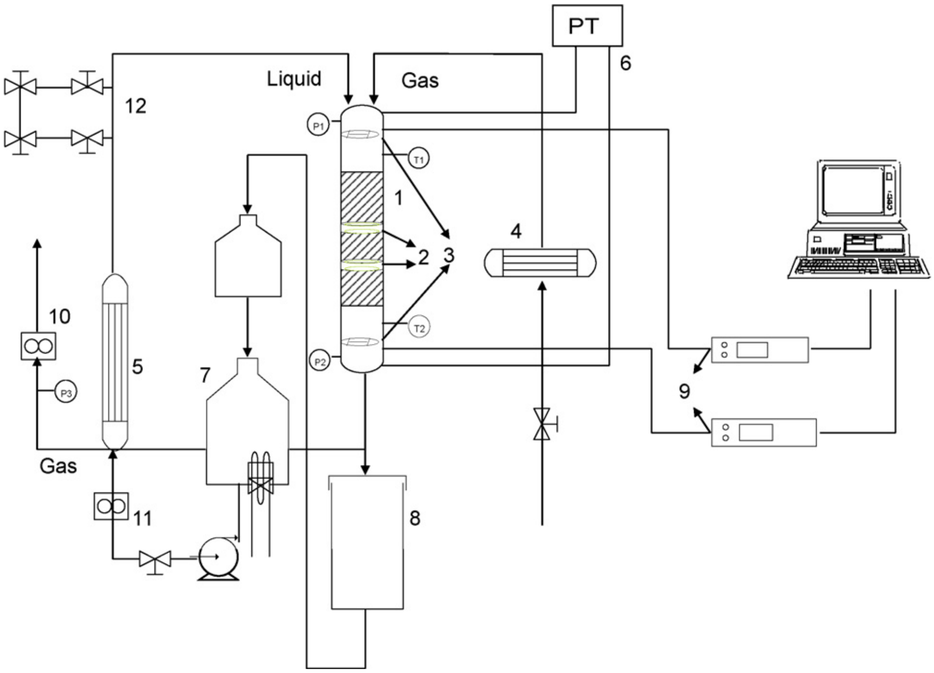

Fig. 1. Trickle-bed experimental setup.

Table 1
Properties of aqueous CTAB and CTAB–CMC solutions at high temperatures and pressures

Temperature (°C)	ρ_{CTAB}[a] (kg/m^3)	μ_{CTAB}[b] $\times 10^4$ (kg/m s)	σ_{CTAB} (kg/s^2)	$k_{CTAB-CMC}$ (kg/m s^{2-n})	$n_{CTAB-CMC}$	$\sigma_{CTAB-CMC}$ (kg/s^2)	k_{CMC} (kg/m s^{2-n})	n_{CMC}	σ_{CMC} (kg/s^2)
25	997.2	8.9	0.050	0.21	0.63	0.052	0.072	0.67	0.056
50	988.2	5.4	0.037	0.14	0.62	0.048	0.041	0.66	0.054
90	964.7	3.1	0.032	0.12	0.61	0.045	0.033	0.66	0.051

[a] Estimated to be close to the values for water.

[b] [26].

state operating temperature was reached along the bed after the reactor was systematically and preventively operated under pulsing flow regime to ensure full bed-wetting.

An electrical conductance technique using ring electrodes was employed for the identification of regime transition and for the investigation of the pulse characteristics as detailed elsewhere [19]. The two electrical conductance probes were mounted in the middle of the reactor, a distance of 0.245 m apart from each other. Each probe was connected to a lock-in amplifier to acquire the output signal. After amplification, the signals were transmitted to a computer by means of a data acquisition system. Identification of flow regime transition was carried out using a moment method [21]. Pulse frequency, f_p, was determined by counting the number of maxima of the conductance trace and dividing by the pulse period for a selected portion of the conductance trace. Pulse velocity, V_p, was determined by dividing the inter-electrode distance by the time delay of maximum cross-correlation between signals.

The two-phase pressure drop was measured with a differential pressure transducer connected to the top and bottom of the packed bed. For liquid holdup measurements, the Aris's double-detection tracer response method was implemented. Two electric conductivity probes – one at the top and another at the bottom of the column – were used. The plug flow with axial dispersion (PD) model was used to determine the liquid holdup (ε_L) by applying a non-linear least squares fitting where the convolution method was used for a time-domain analysis of the non-ideal pulse tracer response data.

3. Results and discussion

3.1. Temperature and pressure evolution of the transition between trickling and foaming-pulsing

Flow regimes in a TBR emerge due to the interaction between phases, which depend on fluid flow rates and physical properties as well as on reactor and particle geometrical features. As for coalescing systems, a low interaction regime, referred to as trickle flow regime at low fluid throughputs, and high interaction regimes at either or both high gas and liquid throughputs exist also for foaming systems. In Fig. 2a and b, the transition boundary from trickle flow to foaming-pulsing flow is plotted as a function of the superficial gas and liquid velocities, the reactor pressure and temperature for the air–CTAB/water and the air–CTAB–CMC/water systems. The observed results are compared, respectively, with the air–water (Fig. 2c) and air–CMC/water (Fig. 2d) systems taken from Ref. [19]. Note that the transition was referred to the displacement from trickling to foaming-pulsing flow, and not from trickling to pulsing flow, as it was difficult to distinguish the pulsing flow regime from the foaming-pulsing flow regime. This was due to a systematic presence of foams, which was recognizable by the larger fluctua-

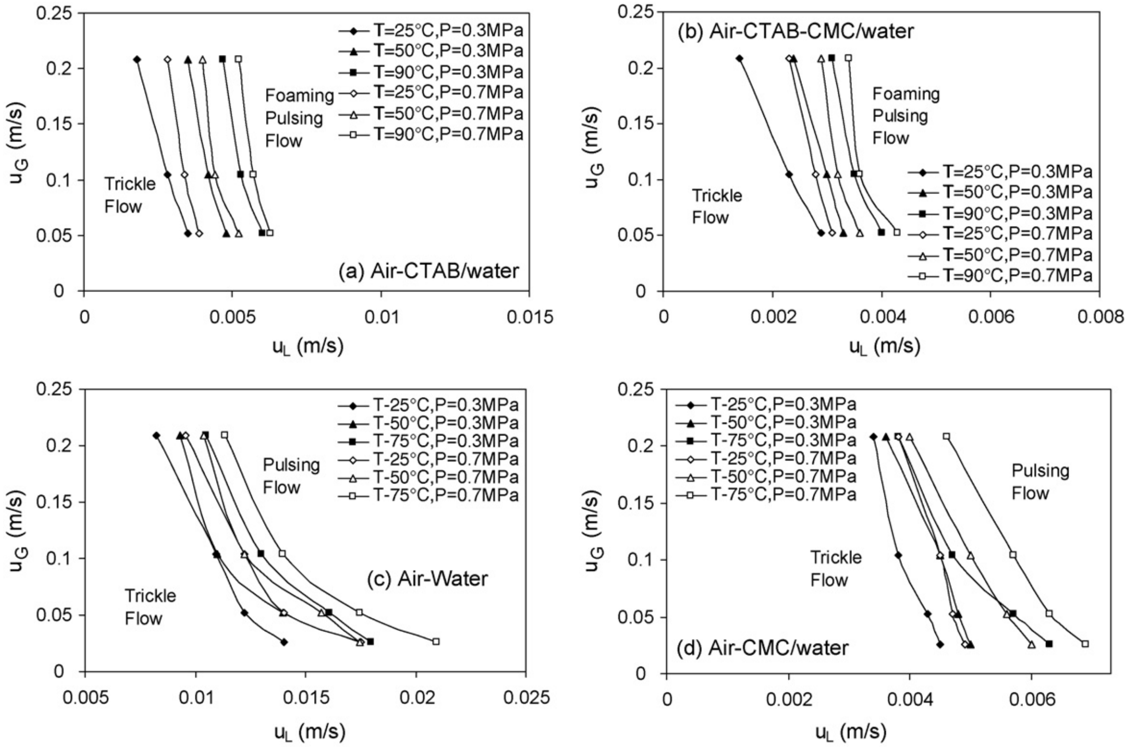

Fig. 2. Influence of pressure and temperature on the transition boundary between trickle and foaming-pulsing flow regimes for the (a) air–CTAB/water, (b) air–CTAB–CMC/water, (c) air–water, and (d) air–CMC/water systems.

tions in the acquired signals in comparison with the non-foaming systems.

The pronounced influence of both reactor pressure and temperature on the transition is illustrated in Fig. 2a and b for both systems. At ambient temperature and constant superficial gas velocity, there is a shift of the transition line towards higher liquid velocities with an increase in reactor pressure. This tendency is classical and is reminiscent of the enlargement of the trickle flow domain as observed for non-foaming Newtonian and non-Newtonian liquids [19] and where the interpretation that a higher liquid volumetric flux is required at elevated pressure to initiate pulse formation is widely accepted. It should be noted that at a given superficial gas velocity, the transition takes place at a much lower superficial liquid velocity for foaming systems than for non-foaming systems (Fig. 2a–d) despite minute changes in the physicochemical properties of the liquids by the introduction of CTAB (Table 1). This is explained as due to early foam formation at lower liquid fluxes as a result of increased pressures; the foams being characterized by lower liquid holdups [4].

Similar to the effect of pressure, the shift towards higher liquid velocities was observed at constant superficial gas velocity with an increase in reactor temperature at constant pressure (Fig. 2a and b). Foam stability is very likely lessened with increased temperatures due to the viscosity decrease of the liquid. This promotes the liquid on the bubble surface to drain faster and to yield unstable bubbles the higher the temperature at given gas and liquid volumetric fluxes. In addition, the resisting forces acting on the liquid phase such as the surface tension force and the liquid shear stress (via viscosity) are weakened with temperature. This causes a decrease in the amount of liquid held within the bed as noted in the behavior of coalescing systems (Fig. 2c and d). Therefore, a higher liquid flow rate is required for the emergence of pulses in the case of foaming systems the higher the temperature. Furthermore, at constant pressure, the effect of temperature on the transition boundary is more pronounced for the foaming systems (Fig. 2a and b) in comparison to non-foaming systems (Fig. 2c and d). Experimentally obtained data for the transition lines for air–CTAB and air–CMC–CTAB systems for all temperature and pressure levels were plotted on the Charpentier and Favier flow regime diagram [22,23] (Fig. 3).

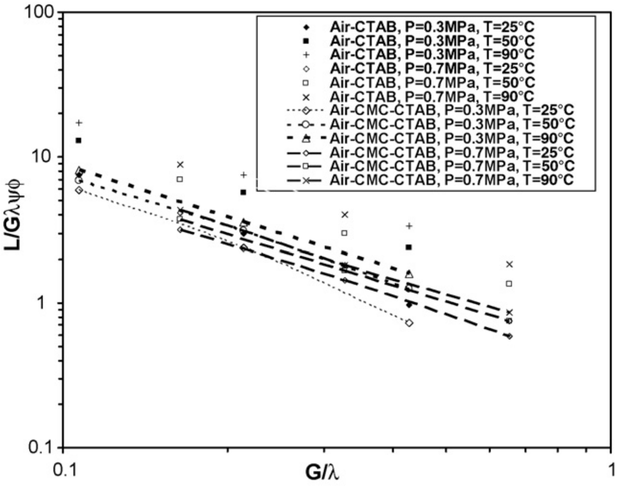

Fig. 3. Plots of experimental trickle-to-foaming-pulsing transition data for air–CTAB and air–CMC–CTAB systems for all temperature and pressure levels on the modified Charpentier and Favier flow regime diagram [23].

Figs. 4 and 5 show the effect of reactor temperature, pressure and superficial gas velocity on the liquid holdup and the two-

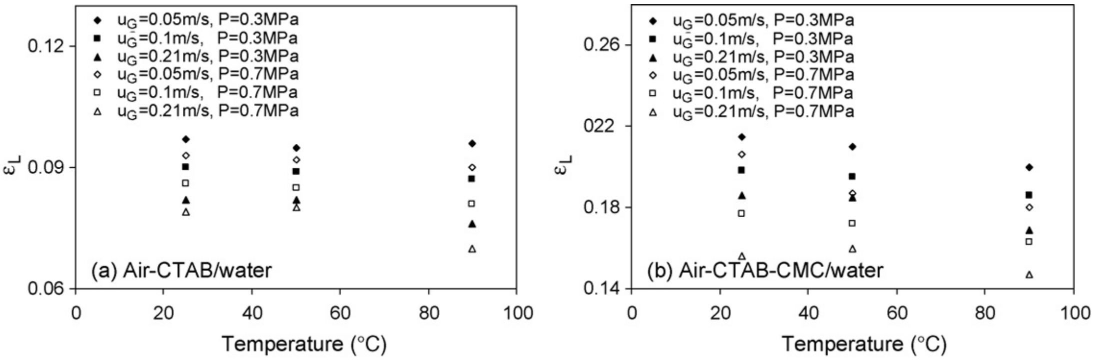

Fig. 4. Effect of temperature, pressure and superficial gas velocity on liquid holdup at trickle-to-foaming-pulsing transition points. (a) air–CTAB/water and (b) air–CTAB–CMC/water.

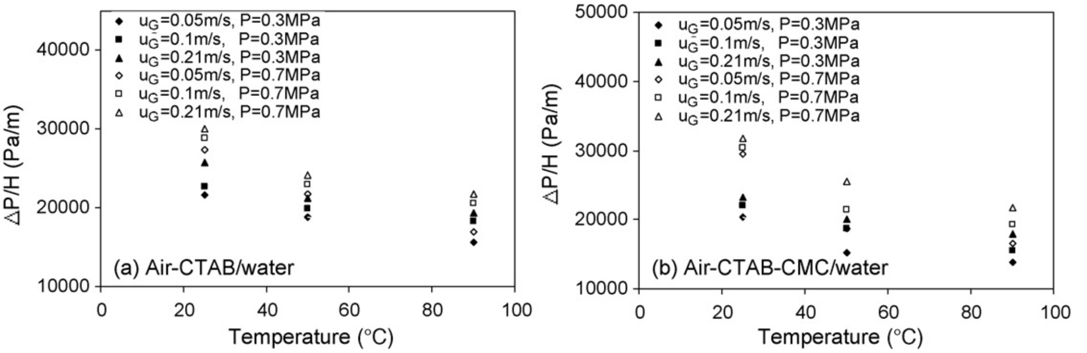

Fig. 5. Effect of temperature, pressure and superficial gas velocity on pressure drop at trickle-to-foaming-pulsing transition points. (a) air–CTAB/water and (b) air–CTAB–CMC/water.

phase pressure drop at the trickle-to-foaming-pulsing transition points: air–CTAB/water (Figs. 4a and 5a) and air–CTAB–CMC/water (Figs. 4b and 5b) systems. For the air–CTAB–CMC/water, the liquid holdup values (ε_L) are larger by ca. a factor two with respect to those corresponding to the air–CTAB/water system. However, the ranges for the transition pressure drops are almost coincident for both systems, typically between 15 and 30 kPa/m. There is a tendency for the transition liquid holdup to decrease especially in the higher temperatures region for both systems and regardless of superficial gas velocity and pressure. This liquid holdup tendency is similar to the one reported for the corresponding coalescing systems by Aydin et al. [24]. The transition pressure drops monotonically decrease with temperature over the whole range of temperatures. Both trends can be related to the liquid viscosity greater sensitivity to temperature, which amongst the physical gas and liquid properties, is the one that experiences the largest reduction when temperature rises from ambient to 90 °C. The pressure drop decreases with temperature in comparable proportions for both systems. Here again, increased temperatures are likely to weaken the frictional forces at the gas–liquid and liquid–solid interfaces as well as liquid surface tension forces resulting in less resistance to flow.

At constant temperature, the transition liquid holdup diminishes with increasing pressure and superficial gas velocity for both systems (Fig. 4). Such liquid holdup reduction is mirrored by the simultaneous increase of the transition pressure drops with increasing gas superficial velocity and/or pressure (Fig. 5). For these experiments, it was observed that the amount of foam produced with increasing pressure is more pronounced for air–CTAB–CMC/water system than that for air–CTAB/water system. This could be intuited from Fig. 4b where a more pronounced fall off of transition liquid holdup with increasing pressure and/or superficial gas velocity takes place. However, the transition pressure drop rise is more sensitive to increasing pressure than to increasing gas superficial velocity (Fig. 4b).

3.2. Liquid holdup and two-phase pressure drop

The effect of reactor temperature, pressure and superficial gas and liquid velocities on liquid holdup for air–CTAB/water and air–water systems is illustrated in Fig. 6a and b and Fig. 6c and d, respectively. These plots include the holdup variations extending from trickle flow to foaming-pulsing flow regime. Similar to the air–water system, liquid holdup increases with superficial liquid velocity for the air–CTAB/water system. As expected, liquid holdup values for the foaming system are much lower than for the non-foaming system for equal fluid volumetric fluxes, and temperature and pressure. Furthermore, liquid holdup increases only slightly for the foaming system (Fig. 6a) in comparison with the non-foaming system (Fig. 6c) over a comparable liquid velocity range. Increasing reactor pressure and superficial gas velocity causes lower liquid holdups. The effect of superficial gas velocity and pressure is drastic for all superficial liquid velocities for the air–CTAB/water system, whereas it is more pronounced for the air–water system only at the higher superficial liquid velocities.

Liquid holdup decreases with increasing temperature for given pressure, and superficial liquid and gas velocities. For the same reasons outlined earlier, the dependence of liquid holdup to temperature is in qualitative agreement with that highlighted on the transition liquid holdup data. The air–CTAB/water system exhibits a remarkable knockdown effect by temperature over the whole superficial liquid velocities (Fig. 6b) whereas the air–water system is sensitive to temperature only in the high liquid throughput region (Fig. 6d).

Fig. 7a–d shows the effect of temperature, pressure, and superficial liquid and gas velocities on liquid holdup for air–CTAB–CMC/water and air–CMC/water systems. At constant gas and liquid velocities and temperature, liquid holdup decreases with increasing pressures for the air–CTAB–CMC/water system similarly to the air–CTAB/water system. However, the effect of pressure for the non-Newtonian system is more visible at high superficial

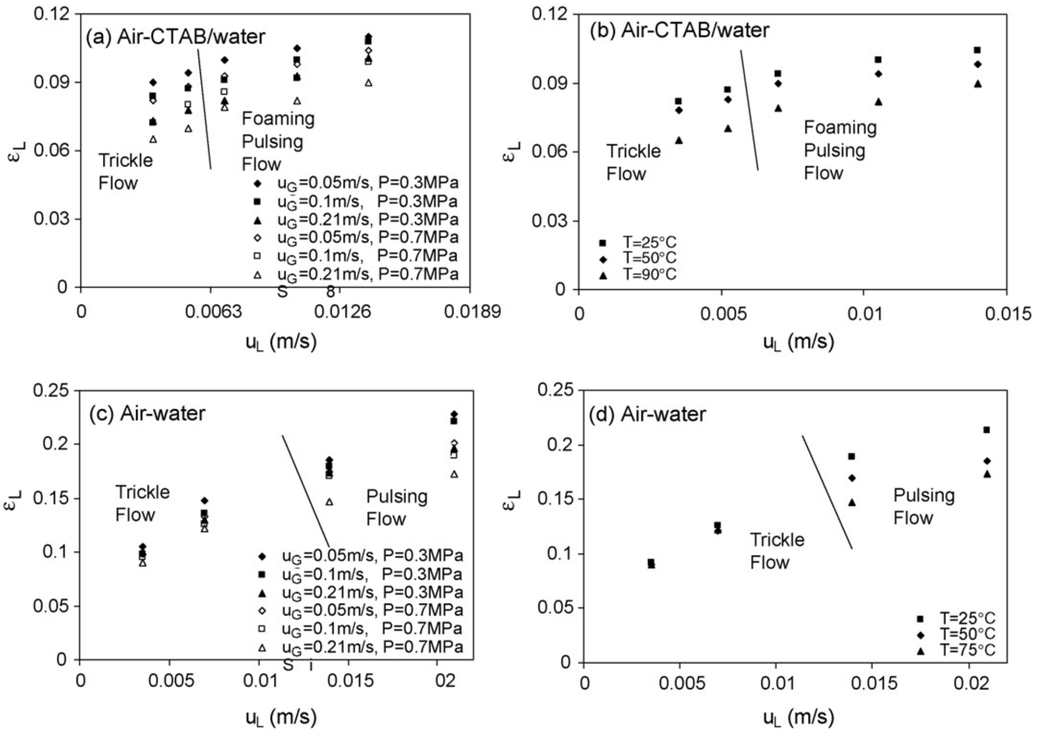

Fig. 6. Influence of pressure and superficial gas velocity (a and c) and temperature (b and d) on liquid holdup for air–CTAB/water and air–water systems.

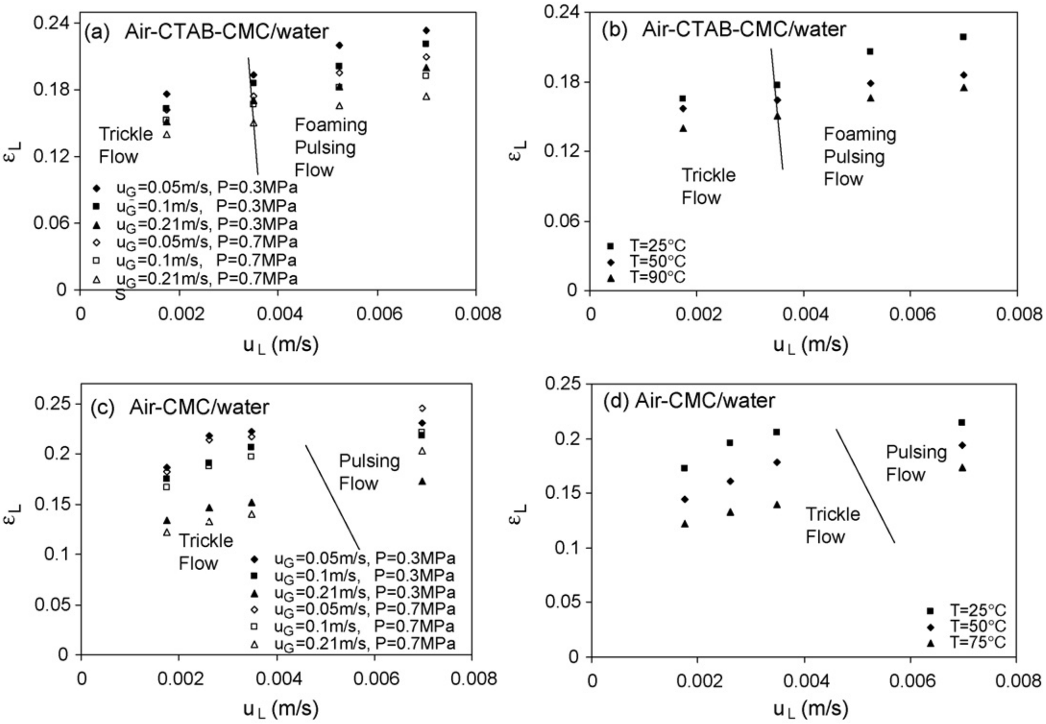

Fig. 7. Influence of pressure and superficial gas velocity (a and c) and temperature (b and d) on liquid holdup for air–CTAB–CMC/water and air–CMC/water systems.

liquid velocity. As expected, the liquid holdup values are larger (Fig. 7a) than those of the air–CTAB/water system (Fig. 6a) due to the viscosity factor. There is a remarkable difference between the effect of pressure and temperature on the liquid holdup for air–CTAB–CMC/water (Fig. 7a and b) and air–CMC/water (Fig. 7c and d) systems. The effect of pressure is more pronounced for the former (Fig. 7a and c) whereas the effect of temperature is more significant for the latter (Fig. 7b and d).

Figs. 8 and 9 show the effect of temperature and pressure on two-phase pressure drop at various superficial liquid and gas velocities for the Newtonian and the non-Newtonian foaming liquids, respectively. At constant superficial liquid and gas velocities, pressure drop decreases with increasing temperature and with decreasing pressure for the same reasons as the pressure drop behavior at the transition point discussed in Fig. 5. As seen in Fig. 8b, the effect of superficial liquid velocity on the pressure drop is very significant. The onset of foaming which is characterized by the interaction between gas and liquid phases is more favorable at higher liquid throughputs occasioning higher pressure drops.

3.3. *Pulse frequency and velocity*

Fig. 10a and b shows the effect of temperature, pressure and superficial liquid velocity on the pulse frequency for the Newtonian and the non-Newtonian foaming systems, respectively. At the same operating conditions, the pulse frequency for the air–CTAB–CMC/water system is higher in comparison to the air–CTAB/water system. This could ascribe to the weaker foaming behavior observed for the Newtonian liquid. The pulse frequency increases with temperature and pressure with a more noticeable incidence from the pressure factor for both systems. At constant temperature and pressure, the pulse frequency also increases with the superficial liquid velocity.

A second basic characteristic of foaming-pulsing flow regime is the pulse velocity which was determined at elevated pressure and temperature as explained in Section 2. Fig. 11a and b illustrates that the pulse velocity increases with temperature at constant superficial liquid velocity and pressure due to a decrease in dynamic liquid viscosity and an increase in interstitial liquid velocity. For the air–CTAB/water system (Fig. 11a),

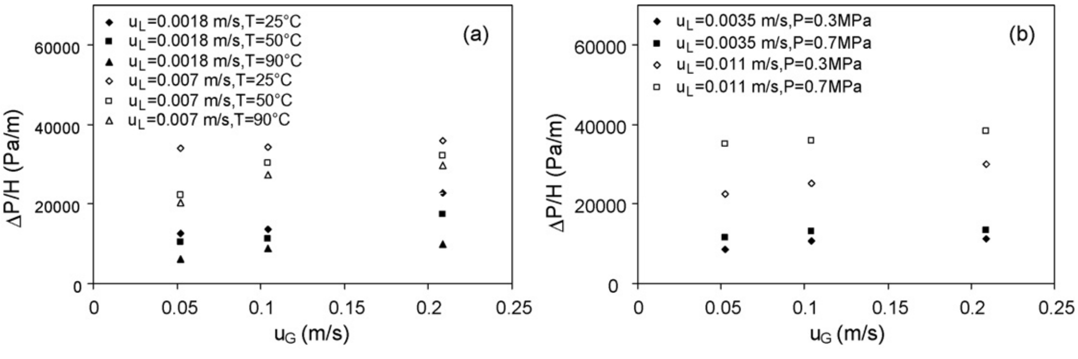

Fig. 8. Influence of temperature (a) and pressure and superficial gas velocity (b) on pressure drop for air–CTAB/water system.

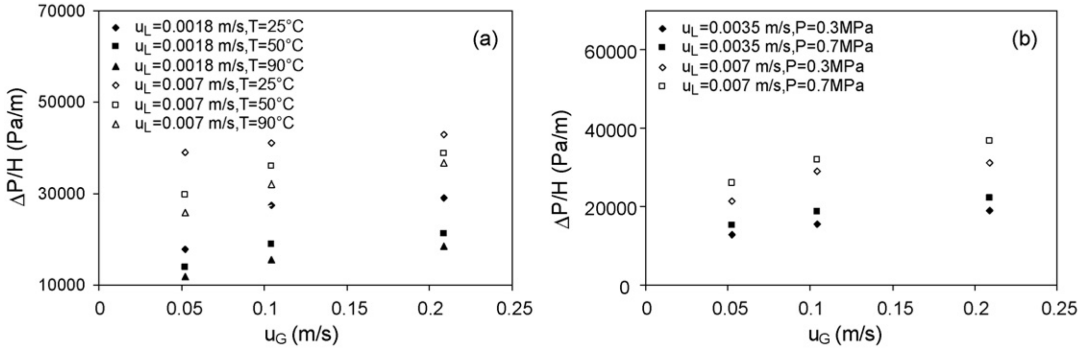

Fig. 9. Influence of temperature (a) and pressure and superficial gas velocity (b) on pressure drop for air–CTAB–CMC/water system.

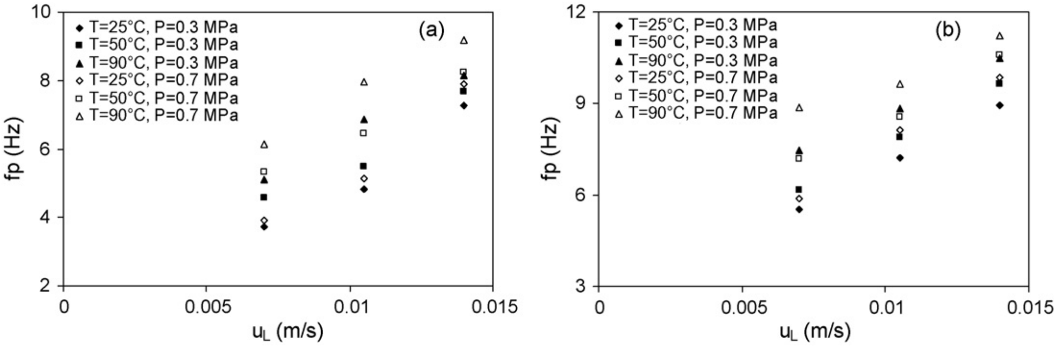

Fig. 10. Effect of temperature, pressure and superficial liquid velocity on pulse frequency for (a) air–CTAB/water and (b) air–CTAB–CMC/water systems. $u_G = 0.21$ m/s.

larger values of pulse velocities were reached in comparison to the air–CTAB–CMC/water system (Fig. 11b) for the same gas and liquid superficial velocities. The gradual increase of the pulse velocity with temperature is similar for Newtonian or non-Newtonian foaming systems. As seen in Fig. 11, the effect of temperature and pressure is more pronounced for foaming systems. The pulse velocity decreases with the increasingly pressure for both systems in accordance with Burghardt et al. [25] and Aydin

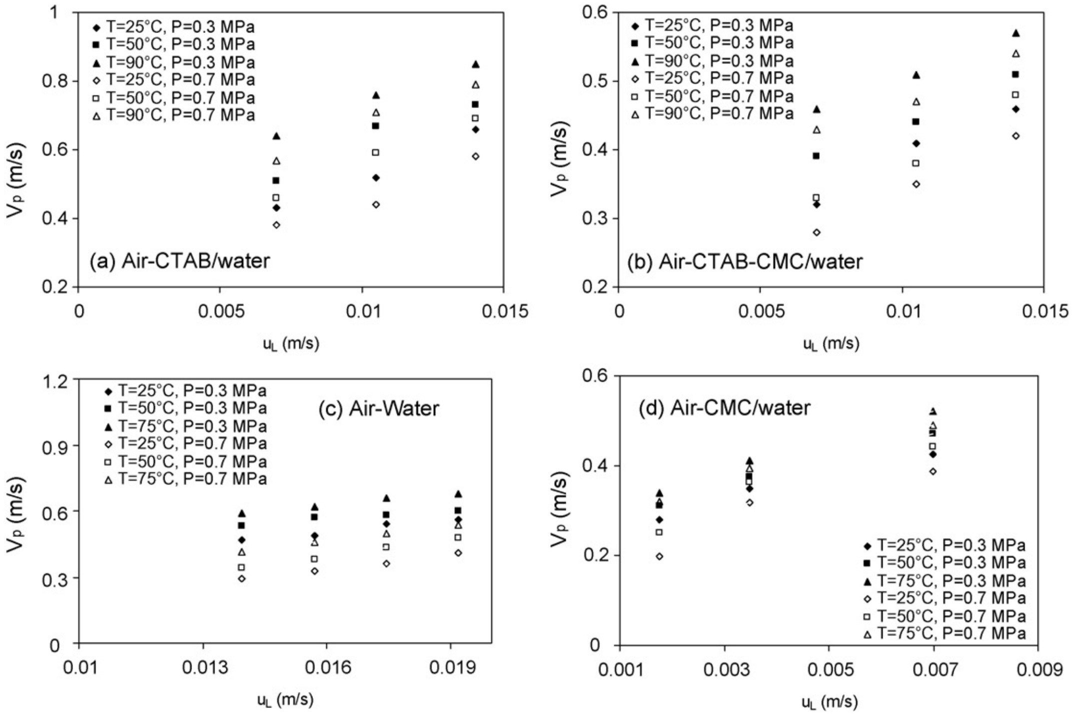

Fig. 11. Influence of temperature, pressure and superficial liquid velocity on pulse velocity for (a) air–CTAB/water, (b) air–CTAB–CMC/water, (c) air–water, and (d) air–CMC/water systems. $u_G = 0.21$ m/s.

and Larachi [19] experimental findings for the coalescing systems.

Fig. 11c and d shows the effect of temperature and pressure on the pulse velocity for Newtonian and non-Newtonian coalescing systems without CTAB addition. At nearly identical liquid and gas superficial velocities, the pulse velocities for the Newtonian coalescing systems (no CTAB) are lower than their Newtonian foaming counterparts (Fig. 11a and c), whereas, non-Newtonian liquids seem to develop an opposite trend (Fig. 11b and d). No explanation can be presented at this moment to interpret these trends.

4. Conclusion

In this study, the effects of moderate temperature and pressure on the hydrodynamics of TBRs were discussed for Newtonian and non-Newtonian foaming systems. The experimental observations were compared with Newtonian and non-Newtonian coalescing systems. The following conclusions were drawn:

- At constant elevated temperature, pressure and superficial gas velocity, the trickle-to-foaming-pulsing flow transition boundary was observed at lower superficial liquid velocity for foaming systems. The transition boundary shifted towards higher gas and liquid superficial velocities with increasingly temperatures and pressures.
- At non-ambient conditions, the liquid holdup for foaming systems was lower than for coalescing systems and pressure drop was higher for foaming systems as known to be the case in room temperature and atmospheric pressure.
- Pulse frequency was an increasing function of temperature and pressure, whereas pulse velocity increased with temperature and decreased with pressure.

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