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Hydrodynamic Properties Investigation of Ebullated Bed Reactor Using Non-Newtonian Liquid

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HIGHLIGHTS

- The effect of non-Newtonian behavior on minimum fluidization velocity, phase hold-up, and bubble diameter in such a reactor was investigated.
- The effect of gas and velocity and reflux ratio was considered.
- Increasing the gas velocity leads to a lower liquefaction velocity. It also decreases with the intensification of non-Newtonian behavior. An increase in the recycle ratio leads to a decrease in the minimum liquefaction velocity.
- The size of the bubbles increases with gas velocity and PMC concentration, while it decreases with the increase in the gas velocity and the recycle ratio.
- The increase in gas hold-up coincided with increased gas and liquid velocities and apparent viscosity.

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1. Introduction

The ebullated-bed reactors (EBRs) are the pinnacle of hydro-processing reactor development. It is used in the hydroprocessing process for feedstock contains high asphalting and metal feedstocks. The bed design allows the exiting solids to flow inside the bed without accumulating, clogging, or increasing pressure drop and provides good mixing between the liquid and particles, similar to that of a fully back-mixed reactor. The essential feature of ebullated-bed reactors is the capacity to intermittently add or even withdraw the catalyst to this type of reactor without disrupting the operation. [1]. The ebullated-bed method was required to deal with the most difficult feeds, such as vacuum residues and heavy oils containing high asphaltenes, metals, and sediments. [2].

The capability of predicting the important characteristics of a three-phase gas-liquid-solid system, particularly the hydrodynamic parameters (e.g., bed expansion behavior, phase hold-ups, and pressure drop), mixing individual phases, and heat 1205

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ABSTRACT

The importance of using EBR has been renewed recently due to the sharp increase in heavy feedstocks sent to refineries and the hydrocracking process. Most of these feedstocks have a non-Newtonian behavior. The performance of this type of reactor using non-Newtonian liquid is complicated and has not been covered well yet. Hence, the present work is devoted to elucidating the effect of the non-Newtonian behavior of fluid on the hydrodynamic properties of a threephase (gas-liquid-solid) reactor under operating conditions of different values of gas velocity (2, 4, 6) cm/sec, liquid velocity (0.9, 1.39, 1.8, 2.3) cm/sec, and recycle ratio (1.5, 2, 2.5). The study observed the effect of non-Newtonian behavior using polymethyl Cellulose (PMC) at different concentrations (0.1, 0.2, 0.3, and 0.4) wt%. The pressure gradient method was used to elucidate the minimum liquid fluidization velocity and to estimate hold up, while the imaging method was used to measure the bubble's size. The results showed that the higher the gas velocity, the lower the minimum liquid fluidization velocity. As the intensity of the non-Newtonian behavior increased, gas velocity showed the opposite effect. The results also showed that increasing the velocity of liquid and gas and the intensity of the non-Newtonian increase the gas hold-up. The bubbles characteristics, represented by bubble size results, show that small bubbles appear at low gas velocities, and these bubbles collapse as gas and liquid velocities increase as well as liquid viscosity.

and mass transfer characteristics, is critical for successful design and operation. [3]. The expansion of the bed is significant in choosing the system's size, but hold-ups of phases are important in the performance of mixing and the overall system [4]. When measuring the hydrodynamics of a fluidized bed for chemical processes, the minimum fluidization velocity is one of the most important factors [5, 6]. The minimum liquid fluidization velocity, U_{lmf} , is the superficial liquid velocity at which the bed becomes fluidized at a specific superficial gas velocity in a three-phase fluidized system. Hence, above the U_{lmf} , all three gas, liquid, and solid phases are mixed very well [7].

The hydrodynamic properties of this type of reactor have been studied by many workers. Ruiz et al. [8] investigate the effect of high temperature and pressure on these properties. In their study, the temperature ranged from ambit (20 °C) to high temperature (100 °C), and pressure ranged between 5.6 and 15.6 MPa. These circumstances were the same as that employed in hydroprocessing using ebullated-bed reactors. Their study showed that the porosity of the bed displays an increase in its values by increasing the viscosity and liquid velocity at high pressure and ambit temperature. In contrast, at high temperatures (100°C), the bed expansion increases with gas velocity. Indeed, the bed porosity values decreased with increasing temperature. They also compared their measurement values of bed expansion and porosity with some analytical correlations and models. Their study also included the effect of temperature on the region of bubble flow. They proposed an empirical correlation at high pressure explaining the transition of bubble flow from dispersed region to coalescence region. Cressman conducted experiments using the radioactive tracer method. They also developed a hydrodynamic model combining the performance of EB with the chemical reactions [1]. Abed et al. [9] studied the hydrodynamics properties of EBRs using air, water, and steel particle system using the pressure gradient method and residence time distribution (RTD) technique for estimating individual impedances and lab-scale elastic bed column dispersion coefficients. Also, the hydraulic efficiency of the system (HEF) was studied. Their results clarified that the internal liquid recycling ratio, which EBRs characterized, affected the individual inactivation and dispersal coefficients. Empirical correlations have been improved to predict phase hold-up and dispersion coefficients accurately. Abed et al. [10] studied the hydrodynamics of flow-bed reactors in a cold model using air, Magnesium Sulfate solution, and a solid catalyst particles system. They worked on the factorial method to study the effect of operating variables on individual inactivation properties and bubble properties. Using the pressure gradient method to determine the porosity and bed layer along the column, they used the imaging method to get some images of the gas bubbles. They developed empirical correlations to predict the bed porosity phase with a good accuracy value. The results displayed that the internal fluid reflux rate was characterized by ebullated bed reactors having a predominant effect on bubble size.

Depending on the literature mentioned above, many researchers have studied the hydrodynamics and kinetics of such a reactor. However, the study of non-Newtonian behavior in such a reactor is not well covered. Accordingly, this study aims to clarify the hydrodynamic properties of the ebulliated reactor using non-Newtonian liquid. The study also aims to show the effect of different operating conditions (liquid and gas superficial velocities), recycling ratio (R) on the hydrodynamic properties of the affected reactor, involving minimum liquid fluidization velocity, bubble diameter, and hold up of gas, liquid and solid. Empirical correlations of the hydrodynamic parameters as a function of the studied operating variables were also estimated.

2. Experimental Work

2.1 Materials

2.1.1 liquid phase

The liquid phase used in this work was poly methylcellulose (PMC) with four different concentrations (0.1, 0.2, 0.3, 0.4) by weight. Methylcellulose (or methyl cellulose) (USA product with a purity of 99.9%) is a chemical compound derived from cellulose, a white hydrophilic powder in pure form. It is non-ionic polymers able to be mixed with other polymers (like polyvinyl alcohol (PVA)), salt, and different ingredients. The PMC solution is a non-Newtonian liquid and is characterized by the use of a viscometer. The rheological properties of the prepared solution are shown in Table 1.

Contraction, wt%	τ0	n	K(cp)	Density (g/cm3)
0.1	1.56	0.92	56.3	1.01
0.2	2.14	0.76	307	1.02
0.3	8.38	0.45	506	1.03
0.4	14.5	0.29	497	1.04

Table 1: Characteristics of a liquid substance used in PMC

2.1.2 Solid phase

The CoMo/Al₂O₃ oxide used in this work is CoMo/Al₂O₃. This product does not contain oxygen, has a large proportion of steadily increasing demand for environmental indicators, and is more stable and miscible with environmentally-friendly engines.

2.1.3 Gas phase

Compressed air was used for this study. The physical properties are density at 20°C (1.2 kg/m³) and viscosity at 20°C (18.36*10⁻⁶ Pa.s).

2.2 Sample Preparations

To know the behavior of residue oil, which is the feed of the ebullated bed reactor, residue oil supplied from AL Dora refinery was used. Their rheological behavior was tested using a viscometer (BROOK FIELD DV3, USA). The results show that

the residue oil has non-Newtonian behavior with Herschel-Buckley model. Hence, PMC has used its solution to show the same non-Newtonian behavior. Table (1) (case: n<1) shows that the fluid exhibits shear-thinning power-law (pseudo-plastic) properties [11].

Four different concentrations (0.1, 0.2, 03, 0.4) wt% were prepared. Their rheological properties were tested using a viscometer. The viscometer (BROOK FIELD DV3, USA) showed the non-Newtonian behavior of the Herschel-Buckley fluid model, as shown in Equation 1:

$$\tau = \tau_0 + K(\gamma)^n \tag{1}$$

The effective average shear rate must be known to calculate the apparent viscosity of the prepared non-Newtonian liquid in the ebullated bed reactor. Therefore, some researchers proposed a correlation to estimate the shear rate. One of these correlations is that proposed by Nishikawa [12] for bubble column reactors. According to their model, the average shear rate is correlated to the superficial gas velocity by the following Equation 2:

$$\gamma = 5000 u_q \tag{2}$$

Applying the superficial gas velocity range used in this work, the effective shear rate inside the bubble column lies in the range of (100-300) 1/s, as shown in Figure 1.



Figure 1: Effective shear rates (1/s) with Viscosity (cp)

2.3 Experimental Setup

The experimental setup used in this work is clarified in Figure 2 (a and b). It consists of an external column with a diameter of 82 mm and a thickness of 5 mm made of acrylic (frosted glass) with a height of 2.0 m. In addition, a fixed amount of solid particles was added to the column with a diameter of 2.5 mm. The feeding tank was filled with 40 liters in two stages. The first stage was tap water. The second stage was poly methylcellulose with concentrations of (0.1, 0.2, 0.3, and 0.4 wt %).

The experiments of this study were carried out at varied liquid velocities. The gas was pumped at different velocities. The liquid and air were entered from the column's bottom part through the pre-mixing area under the distributor network. This area contains plastic rings and ceramic to increase liquid and gas distribution. Mainly, water was introduced as the initial stage, after which the liquid feed pump (P1) was started. Then, the column was turned on the recycling pump (P2) quickly at different recycle ratios (1.5, 2, 2.5) for the recycled liquid recovered to the recycling beaker near the top part of the reactor via an internal return line of the uniform diameter of 25 mm located inside the reactor vessel with the recirculation pump (F3). A compressor was used to source the column with the air at a required flow rate through the flow meter (F2). The solid particles in the layer are lifted by the higher flow velocity of gas and liquid that keeps the particles of the "blown layer" in random motion. All the pressure taps were linked to a pressure gauge (Bourdon gauge, with 4 in and gage measure of 0- 200 mbar, Germany) to measure the local pressure inside the column.

The experiment was given enough time (15 minutes) to reach a steady state. Then, each run was repeated (2-3) times, and the average values were adopted.



Figure 2: The experimental rig: (a) photograph view, (b) schematic diagram

2.4 Measurement Techniques

The techniques which are used to measure the parameters of minimum fluidization velocity and phase hold-ups are:

2.4.1 Minimum fluidization velocity

Fluidization minimum can be defined as the minimum superficial velocity at which the upward pull force equals the downward force. It is determined from the plot of pressure drop across the bed versus fluid velocity. Continuously the measured pressure drop across the layer with the velocity of the liquid under different gas velocity conditions, recycle ratio, and PMC concentrations. In all these numbers, the pressure drop increases with velocity (first region) and eventually reaches a constant pressure drop (second region). This first zone is called the fixed bed zone, and the second is the fluidized bed zone. The point of intersection of the increasing portion of the curve and the constant region represents the minimum fluidization velocity, as shown in Figure 3.



Figure 3: Dimensionless pressure drop versus superficial velocity [13]

2.4.2 Phase hold-up measurements

The hold-ups (g, l, and s) were detected by pressure drop measurement. The pressure drop is related directly to the density of each phase, as follows:

$$\Delta P = gH_s \left(\rho_g \mathcal{E}_g + \rho_l \mathcal{E}_l + \rho_s \mathcal{E}_s \right) \tag{3}$$

The solid hold-up can be calculated based upon the observed bed expansion and the known solid loading of the bed:

$$\mathcal{E}_{s} = M_{s} / \rho_{s} A_{R} H_{s} \tag{4}$$

And since the existing phases are the gas, liquid, and particles, then

$$\mathcal{E}_{g} + \mathcal{E}_{l} + \mathcal{E}_{s} = 1 \tag{5}$$

Equation 4 requires a constant solid hold-up across the expanded bed, which may not be correct. However, the interface difference between the free solid and ebullated bed regions may be discerned. For the three-phase hold-ups at various heights along the column, Equations 3, 4, and 54 were solved to get Equation 5 [14].

$$\varepsilon_G = \frac{\left(\frac{\Delta P}{\Delta Z}\right)g^{-1} + \left(\rho_S - \rho_L\right)\varepsilon_S}{\rho_L - \rho_G} \tag{6}$$

 (ρ_L) is liquid density, (ρ_S) is particle density, (ρ_G) is gas density, (P) is the pressure, and Δz is the vertical distance between differential pressure taps (m).

2.4.3 Bubble characteristics

The photographic method was used in this work because of its advantages in measuring the simultaneous bubble velocities, bubble diameters, and gas hold-up. A Dual 12MP Ultra Wide and Wide cameras (Ultra Wide: /2.4 aperture and 120° field of view; Wide: /1.8 aperture, 2x optical zoom out, and digital zoom up to 5x) were used in the experimental work to monitor the bubble characteristics in the bubble column. Images were analyzed using The Ai Adobe Illustrator CC (64 Bit) software to analyze the images and estimate the average bubble diameter.

3. Results and Discussion

3.1 Minimum Liquid Fluidization Velocity (Ulmf)

3.1.1 Effect of gas velocity on minimum fluidization velocity

Figures (S1-S5) show the effect of gas velocity on the minimum fluidization velocity with different recycling ratios and PMC concentration values. It can be seen that U_{lmf} values decreased with the increase of gas velocity. This may be caused by increased gas in the reactor, reducing the column cross-section for the liquid flow. The same trend can be observed for all values of the recycling ratio and PMC concentration. Similar results have been shown by another research [15].

3.1.2 Effect of rheological properties

The differences in the minimum fluidization velocity with different concentrations of PMC solution are shown in figures (S1 –S5). It is detected that the fluidization velocity decreases with increasing PMC concentration. This can be because increasing the concentration of PCM causes an increase in the liquid viscosity, i.e., an increase in the pseudo-plasticity of the fluid. This means an increase in the fluid viscous force that can lead to reduced liquid velocity and minimum fluidization velocity. Miura and Kawase showed a similar effect of pseudo-plasticity on minimum fluidization velocity [15].

3.1.3 Influence of internal reflux ratio (R)

Through the experimental work (Figures [S1-S5]), it is noted that the increase in recycling (R) leads to a decrease in the minimum velocity of liquid fluidization. Since the recycle ratio is the ratio of liquid flow recycled from the column to the liquid flow rate entering the reactor, the increasing recycle ratio means a decrease in the available cross-section of the reactor for the gas to flow due to the increase of the liquid hold up. Hence, that can reduce the minimum liquid fluidization velocity. Therefore, the internal gradient ratio can be considered one of the effective factors related to fluidization.

3.1.4 Empirical correlation

The minimum fluidization velocity was correlated to the following variables (n, K, ρ L, ρ G, μ_{eff} , d_P, ug, and R). The operating variables used are non-Newtonian liquid with different concentrations, different gas velocities, and reflux ratios. A dimensional analysis of the Buckingham π - Theorem was used to derive an empirical equation. A statistical analysis program was used to estimate the correlation coefficient, variance, and average absolute error of the equation and the coefficient and exponents. A comparison between the predicted values and the experimental results is represented in figure (4). The predicted equation is well allied with the experimental values with correlation coefficient R =0.95 and variance 0.9.

$$Re_{mf} = -1.53Re^{0.024} + 0.0013 Ar^{1.23} + 1.87 R^{-0.11}$$
(7)

In which: Ar= Archimedes No $Ar = \frac{d_p^3 g(\rho_p - \rho l)}{\mu_{eff}^2}$ Re= particle Reynolds No $Re = \frac{U_1 d_p \rho_1}{\mu_{eff}}$ $\mu_{eff} = effective viscosity = K(\gamma)^{n-1}$



Figure 4: The experimental results versus the predicted values

3.2 Hold-Up Values

3.2.1 Effect of gas and liquid velocities

The effects of studied variables of superficial gas and liquid velocities, reflux ratio, and non-Newtonian behavior on the gas hold-up are shown in Figures (S6-S11). The figures show that the increase in gas velocity results in the gas hold-up values increasing due to the increase of gas fraction in the column. This agrees with the previously published results [16]. Also, the increase in superficial liquid velocity causes an increase in the gas hold-up. This result contradicts the work obtained by [16], which shows the inverse effect. This can be explained by the effect of high shear force, rusted by increasing liquid velocity, which can lead to forming large bubbles. Since large bubbles have less velocity, increasing the retention time of the bubble in a certain position and, hence, increases gas hold up.

The liquid hold-up values obtained from the experimental work are investigated for their reliance on the operating variables of gas and liquid flow rate, reflux ratio, and the rheological properties of the liquid phases. The results are shown in figures (S12- S16). The figures show that increasing liquid flow rate increases the liquid hold up due to the increase of liquid existence and the high drag forces applied to the solid particles by the increase in liquid velocity. However, the liquid flow rate has larger effects on gas hold up than on liquid hold up. This observation is due to the high porosity, and the flowing liquid, even at high flow rates, cannot eliminate gas bubbles in the column.

On the other hand, the liquid hold-up decreases with increasing gas velocity since that increasing gas velocity means an increase in the fraction of gas bubbles in the column, and hence liquid hold-up decreases. Also, with increasing gas velocity, the mean liquid residence time necessarily decreases due to greater shear at the interface between gas and liquid. This leads to a decrease in the liquid fraction in the column. Therefore, the reduction rate in total liquid hold up is faster at lower gas velocity values than at higher values. These results agree with the work of [16]. While in another research [17], the liquid hold-up variation with gas velocity increasing was not significant.

Through Figure (S17- S20), it can be noticed that, at constant liquid velocity, solid hold-up decreases with the gas velocity increasing. This can be attributed to the larger drag force on a solid particle as the liquid velocity increases, causing an expansion in the solid bed and hence a decrease in the solid hold-up. Likewise, the increase of liquid velocity, for constant gas velocity, will reduce solid hold-up since increasing the liquid's velocity reduces the solid's cohesion and thus reduces the stopping of the solid inside the column. Again, the same trend was observed in the literature.

3.2.2 Effect of rheological properties

The behavior of non-Newtonian liquid in three phases on hold-up is very complex. Therefore, the gas, liquid, and solid holdup dependency on the rheological properties of liquid were investigated using water and four solutions of PMC with concentrations.

The Figures (S6-S20) show that increasing of fluid consistency index from 0.29 to 0.92 leads to an increase in the gas holdup. This is attributed to this increase related to a decrease in liquid viscosity, and it is well known that for viscous liquids, the energy of eddies, as well as the turbulence effects that causing a break the large bubbles to a small size, is reduced while bubble coalescence is promoted. Hence, gas bubbles inside the column have a greater resistance to movement. Therefore, the gas bubbles' residence time increases, increasing the gas hold up.

Indeed, at higher effective shear rates inside the column (100 to 300), 1/s corresponding to the increase of gas velocity from 2 to 4 cm/s to 0.1, 0.2, 0.3, and 0.4w% of PMC, respectively), as shown in Figure (1). The viscosity of the PMC solution drops and lowers with the decrease of PMC concentration. Accordingly, the decrease in the values of the solution viscosity is related to the higher gas hold-up of the PMC solution at the higher values of the gas velocity.

A possible explanation for the fluid accumulation of PMC concentration is that the overall effect of the rheological nature of PMC on fluid suspension depends on the competitive tendency between viscosity and shear anomaly. On the other hand, at constant fluid velocities, the decrease in fluid consistency index causes a decrease in the liquid hold-up. This result is revealed

with the result obtained by [17], who concluded that the increase in the liquid hold-up is attributed to increasing the liquid viscosity to fluid consistency index ratio, which in turn enhances the liquid shear stress.

Also, higher PMC concentration indisputably means higher viscosity and, therefore, higher pseudo-elasticity. In addition, a higher viscosity means an increase in the velocity of the interstitial gas, which can cause the fluid flowing downward to be drawn upwards. On the other hand, the increase in the non-Newtonian anomaly or the shear-thinning decreases the effective viscosity of the PMC aqueous solution and thus increases the suspension of the liquid. Therefore, a higher PMC content in the solution results in higher liquid suspension profiles, regardless of the amount of liquid flowing down the column. Of course, for water flow without PMC content, the liquid suspension has the lowest. This is in agreement with [18-20] who showed that an increase in gas retention in the bubble column is associated with an increase in viscosity and surface tension of the liquid. In contrast, the studies by [21-23] stated that the increase in viscosity is offset by a decrease in gas retention due to a decrease in the critical gas velocity for a complete suspension of solid particles by increasing the viscosity of the liquid and thus reducing gas [24] that the gas remains unaffected by the viscosity of the liquid.

The effect of liquid viscosity on the solid hold-up of solids is shown in previous figures for different initial velocities. To obtain the same overall velocity of the viscous solution, the solid hold-up decreases with an increasing superficial velocity of the liquid system. Still, with increasing viscosity, the solid hold-up increased for all concentrations. This is because the initial velocity dominates over the auxiliary velocity, which traps more solids from the riser. Thus less difficult stopping is observed at the lower viscous solution.

3.2.3 Effect of internal reflux ratio

Through experimental work, the figures show that the internal regression ratio is one of the effective parameters that can be used to control the deceleration rate of the catalyst bed. It should be noted that an increase in recycling (R) leads to a decrease in solid retention. Since the recycling ratio is the ratio of the recycled liquid flow from the column to the liquid flow rate entering the reactor, an increase in the recycling ratio means an increase in the liquid hold up in the column. This results in a higher bubble break up, reducing small bubbles and thus increasing the gas retention within the column.

Also, it can be seen from the figures that when the velocity of the liquid rises, the time for gas exit from the liquid is less, and therefore the gas retention in the recycling line is generally higher. The effect becomes more pronounced with increasing gas velocity. Similar behavior was also reported by [25] in the Amoco cold flow unit. Note that the gas hold-up in the recycling line doubled with the increase of the inlet fluid velocity from (0.9 to 2.3) cm/s. Increasing superficial velocity to reduce the superficial velocity ratio between gas, liquid, and liquid (U_G / U_L), increase the Reynolds number of particles (Re_{L-S}), and increase gas retention in the layer region at higher superficial velocities of the liquid. The gas retention in the layer region in the fusion system increased, most likely due to the increased dissociation of bubbles in the layer. The layer expanded when the U_G / U_L increased due to the increase in the volumetric gas fraction.

3.2.4 Empirical correlation

Gas, liquid, and solid retention values correlate well in the following operating variables (n, Ug, Ul, and R). The operating variables that were used are a non-Newtonian fluid with four different concentrations with different gas velocities (2-6) cm/sec, liquid velocities (0.9- 2.3) cm/sec, and reflux ratios (1.5- 2.5). The dimensional analysis of Buckingham's theory [13] was used to derive an empirical gas

equation:
$$Eg = -2.09 + 2.29n^{-0.01} + 1.25Ug^{0.02} - 1.13Ul^{-0.001} + 0.7R^{0.99}$$
 (8)

Liquid:
$$El = -0.5 + 0.51n^{-0.04} - 0.05Ug^{0.42} + 0.06Ul^{-0.048} + 0.21R^{-0.04}$$
 (9)

Solid:
$$Es = -1.24 + 2.77n^{-0.01} + 0.14Ug^{-0.2} - 1.47Ul^{0.02} + 0.04R^{-0.2}$$
 (10)

Statistical analysis software was used to estimate the correlation coefficient, variance, and mean absolute error to calculate the coefficient and exponents. The experimental results and expected values are compared in Figures 5, 6, and 7. The predicted equation fits well for the experimental values with the correlation coefficient and variance For gas R^2 = 0.85, variance = 0.5, liquid R^2 = 0.83, variance = 0.69 and solid R^2 = 0.8, variance = 0.64.

The velocity of the liquid (U_l) , as shown in equations (8, 9, 10), has little effect on the gas hold-up while it mainly affects the liquid and the solid hold-up. On the other hand, gas velocity (U_g) greatly affects gas hold-up and liquid hold-up and less affects solid hold-up. The solids hold-up depends on the recycling ratio (R) and mainly affects the gas hold-up; the effect of the recycling ratio is less on the liquid hold-up. While the effect of the behavior of the non-Newtonian fluid (n) on the (gas, liquid, and solid) hold-up was almost the same.



Figure 5: Comparison between the experimental results and the predicted values solid hold up



Figure 6: Comparison between the experimental results and the predicted values liquid hold up



Figure 7: Comparison between the experimental results and the predicted values of gas hold-up

3.3 Bubble Characteristics

3.3.1 Effect of gas and liquid velocities

The effects of both gas and liquid superficial velocities on the diameter of the bubble are shown in Figures (S21-24). It can be noticed from these figures that as the volumes of liquid passing through the bed region are caused by increasing liquid flow rates, the tendency of the bubble approach enhances, leading to the likelihood of bubble coalescence. Moreover, gradually these bubbles are larger due to the promotion in the presence of large/coalescing bubbles as a result of the wake effect of large bubbles, which enhances the bubble collision [26].

Indeed, at low gas velocities, small bubbles appear. These bubbles collapse by increasing gas velocities.

In Newtonian fluid systems, most bubbles are small. However, for highly viscous liquids, a bubbling flow system occurred. In this system, small bubbles with diameters (0.7-1.8) cm and almost spherical cap shape bubbles rise upward directly with relatively little interaction with each other. At higher gas velocity (6 cm/s), the large spherical cap bubbles were formed by docking near the waiter and rising in zigzag paths. Based on visual observation, the size of the large spherical bubble ranged between (2.3 - 5.5) cm, depending on the fluid's viscosity.

3.3.2 Effect of PMC concentration on bubbles diameter

The effect of liquid viscosity on bubble diameter is shown in Figures (S21-S24). It is noticed from these figures that the bubble diameter increases with the increase in viscosity of the liquid. This may be attributed to the fact that adding PMC with different concentrations resulted in interfacial phenomena inhibiting bubble coalescence. In other words, the viscosity increase promotes bubble fusion, leading to an increase in bubble diameter. This behavior of bubble diameter is the same as that reported by [27].

The bubbles' behavior in highly viscous fluids or non-Newtonian fluids is relatively different from that of other lowviscosity fluids or Newtonian fluids. The bubbles merging and rupturing of bubbles at the surface results in very small bubbles that accumulate and circulate with the liquid due to their low velocity. These two phenomena result in various bubble size distributions, which are seen in both Newtonian and non-Newtonian fluids. However, in non-Newtonian liquid systems, the large size of bubble formation is strongly enhanced.

3.3.3 Influence of internal reflux ratio (r)

The same mentioned figures indicate the effect of the internal flow ratio on the bubble size. Those figures show that increasing reflux ratio results in decreasing in bubble diameter. This can be explained by as the reflux ratio increases, the intensity of the turbulence also increases, causing high shear stress on the surface of the bubble. These shear forces promote the collapse of bubbles into smaller bubbles. The bubble size stability, as well as bubble break-up, are functions of the bubble interface forces. Shear stress is increased due to higher turbulent intensity resulting in a high centrifugal force that acts outwards on the bubble surface. As this internal centrifugal force exceeds the gas-liquid surface tension force, the bubble break-up is enhanced, and bubble stability is reduced.

4. Conclusion

- 1) Increasing the gas velocity leads to a lower fluidization velocity. It also decreases with the intensification of non-Newtonian behavior. Also, an increase in the recycle ratio causes a decrease in the values of minimum fluidization velocity.
- 2) The increase in gas hold-up is related to an increase in gas and liquid velocities and apparent viscosity.
- 3) Increasing the liquid flow rate increases liquid hold-up values, while they are related inversely to the gas velocity.
- 4) Solid hold-up decreases with increasing gas velocity. Similarly, increasing the velocity of the liquid will reduce the solid suspension. However, with increasing viscosity, it was found that the solid suspension increased for all concentrations. Therefore, an increase in recycling (R) leads to a decrease in solids hold-up.
- 5) The size of the bubbles increases with gas velocity and PMC concentration, while it decreases with gas velocity and recycles ratio increasing.

AR	Archimedes No	Re	particle Reynolds
DP	particle diameter (m)	S	solid
g	gravitational acceleration (m/s2)	UG, UL	gas and liquid superficial velocities (m/s)
G	gas	Ulmin	minimum liquid fluidization velocity
Hs	Solid high (cm)	Δz	the vertical distance between differential pressure taps (m)
L	liquid	EG	Gas hold-up
MS	Solid mass (kg)	EL	Liquid hold-up
Ν	behavior index	ES	Solid hold-up
Р	pressure (Pa)	PG, pL	gas and liquid dynamic viscosity (Pa)
PMC	polymethyl cellulosic	Y	shear rate
R	Reflux Ratio		

Nomenclature

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Author contribution

All authors contributed equally to this work.

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

References

- S. Cressman, C. Boyer, J.J. Colyar, J.M. Schweitzer, and J.C. Viguie, Improvements of ebullated-bed technology for upgrading heavy oils, Oil Gas Sci. Technol., 55 (2000) 397-406. <u>http://dx.doi.org/10.2516/ogst:2000028</u>
- [2] A. Chander, A. Kondo, SC. Big, A. K Dalai, and D. K Fora, Hydrodynamic characteristics of cocurrent upflow and downflow of gas and liquid in a fixed bed reactor, Fuel, 80 (2001) 1043-1053. <u>http://doi:10.1016/S0016-2361(00)00170-8</u>
- [3] H. Al-Sadiqi, B. Gourich, C. Vial, and H. Delmas. Residence time distribution measurements in an external-loop airlift reactor: Study of the hydrodynamics of the liquid circulation induced by the hydrogen bubbles, Chem. Eng. Sci., 66 (2011) 3125-3132. <u>http://doi.org/10.1016/J.CES.2011.02.063</u>
- [4] H. Fogler, Scott. Elements of Chemical Reaction Engineering, 3rd edn. Printice-Hall, Inc, New Jersy 1999.
- [5] F. G. Froment., Kenneth B. Bischoff, and Juray De Wilde. Chemical reactor analysis and design 3rd Edition, New York: Wiley., 2(1990)
- [6] G. R. Caicedo, M. G. Ruiz, J. J. P. Marqués, and J. G. Soler. Minimum fluidization velocities for gas-solid 2D beds, Chem. Eng. Process: Process Intensif., 41 (2002)761-764. <u>http://doi.org/10.1016/S0255-2701(02)00005-3</u>
- [7] Z. Fu, J. Zhu, S.Barghi, Y.Zhao, Z. Luob and C.Duan, Minimum fluidization velocity of binary mixtures of medium particles in the Air Dense medium fluidized bed, Chem. Eng. Sci., 2 (2019)194-201.
- [8] R.S. Ruiz, F. Alonso, J.Ancheyta Pressure and temperature effects on the hydrodynamic characteristics of ebullatedbed systems, Catal. Today, 109 (2005) 205-213. <u>http://doi.org/10.1016/j.cattod.2005.08.019</u>
- [9] M. F. Abid, Z. Y. Shanain, and K. N. Abed. Experimental and analysis study on dispersion of phases in an Ebullated Bed Reactor, Oil Gas Sci. Technol., 74 (2019) 20. <u>http://doi.org/10.2516/ogst/2018103</u>
- [10] M. F. Abid, S. M. Ahmed, H. H. Hasan, D. Al-Mously, and S. Barghi, Hydrodynamic Study of an Ebullated-bed Reactor in the H-oil Process, Iran. J. Sci. Technol. Trans. A: Sci., 43 (2019) 829-838.
- [11] N. J. Alderman, Non-Newtonian Fluids: Guide to Classification and Characteristics, Report number, ESDU Data Item 97034 Affiliation: ESDU International plc, (1997).
- [12] M. Nishikawa, H. Kato, and K. Hashimoto. Heat transfer in aerated tower filled with non-Newtonian liquid, Ind. Eng. chem. process des. Dev., 16 (1977)133-137. <u>http//doi.org/10.1021/i260061a607</u>
- [13] JF. T. Richardson. Sedimentation and fluidization, Trans. Inst. Chem. Eng., 32 (1954) 35-53.
- [14] D. Pjontek, and A. Macchi. Hydrodynamic comparison of spherical and cylindrical particles in a gas-liquid-solid fluidized bed at elevated pressure and high gas holdup conditions, Powder Technol., 253 (2014) 657-676. <u>http://doi.org/10.1016/j.powtec.2013.12.030</u>
- [15] H. Miura, and Y. Kawase; Hydrodynamics and Mass Transfer in Three-Phase Fluidized Beds with Non-Newtonian Fluids, Chem. Eng. Sci., 52 (1997) 4095–4104. <u>http://doi.org/10.1016/S0009-2509(97)00251-0</u>
- [16] S. Venkatachalam, A. Palaniappan, and K. Kandasamy. Hydrodynamic Studies on Three-Phase Combined (Internal & External) Loop Airlift Fluidized Bed Reactor Using Newtonian and non-Newtonian Liquids: Minimum Fluidization Velocity and Liquid Holdup, Int. J. Chem. React., 9 (2011)
- [17] V. Sivakumar, K. Senthilkumar and T. Kannadasan. Prediction of liquid holdup and solid holdup in three-phase fluidized bed: air/newtonian and non-newtonian liquid systems, Chem. Biochem. Eng. Q., 22 (2008) 401-410. <u>http://doi.org/10.2478/v10026-010-0053-7</u>

- [18] V. Elgozali, Linek, M. Fialova, O. Wein, and J. Zahradnık, Influence of viscosity and surface tension on performance of gas-liquid contactors with ejector type gas distributor, Chem. Eng. Sci., 57 (2002) 2987-2994. <u>http://dx.doi.org/10.1016/S0009-2509(02)00165-3</u>
- [19] A. Schumpe, & W.D. Deckwer. Gas holdups, specific interfacial areas, and mass transfer coefficients of aerated carboxymethyl cellulose solutions in a bubble column, Ind. Eng. chem. process des. dev., 21 (1982) 706-711.
- [20] S. Khare, and K. Niranjan. The effect of impeller design on gas hold-up in surfactant containing highly viscous non-Newtonian agitated liquids, Chem. Eng. Process, 41 (2002) 239-249. <u>http://doi.org/10.1016/S0255-2701(01)00139-8</u>
- [21]S. V. Dharwadkar, S. B. Sawant, and J. B. Joshi. Gas hold-up in highly viscous pseudoplastic non-newtonian solutions in three phase sparged reactors, Can. J. Chem. Eng., 65(1987) 406-411. <u>http//doi.org/10.1002/cjce.5450650307</u>
- [22] K.Ho, and W. K. Lee. Circulation liquid velocity, gas holdup and volumetric oxygen transfer coefficient in externalloop airlift reactors, J. Chem. Technol. Biotechnol., 56 (1993)51-58. <u>http://doi.org/10.1002/jctb.280560110</u>
- [23] G. Kelkar, and Y. T. Shah. Gas holdup and backmixing in bubble column with polymer solutions, ChE J., 31 (1985) 700-702. <u>http://doi:10.1002/AIC.690310424</u>
- [24] Molina, A. Contreras, and Y. Chisti. Gas holdup, liquid circulation and mixing behaviour of viscous Newtonian media in a split-cylinder airlift bioreactor. Food Bioprod. Process., 77(1999) 27-32. <u>http://doi.org/10.1205/096030899532222</u>
- [25] W. T. Tang and L. S. Fan. Steady state phenol degradation in a draft-tube, gas-liquid-solid fluidized-bed bioreactor, ChE J., 33 (1987) 239-249. <u>http//</u>doi.org/10.1002/aic.690330210
- [26] M. W. Haque, K. D. P. Nigam, V. K. Srivastava, J. B. Joshi, and K. Viswanathan. Studies on mixing time in bubble columns with pseudoplastic (carboxymethyl) cellulose solutions, Ind. Eng. Chem. Res., 26 (1987) 82-86. <u>http://doi.org/10.1021/ie00061a015</u>