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State of the art

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ABSTRACT

All important studies on the influence of pressure on mass transfer phenomena in gas-liquid systems and reactors are critically reviewed. Points of agreement and conflict will be indicated and discussed.

It is concluded that:

1. The initial bubble size at a single orifice decreases with increasing pressure.
2. The gas phase mass transfer coefficient k_G is inversely proportional with pressure to the power n , where n depends on the mass transfer mechanism.
3. The liquid phase mass transfer coefficient k_L is not influenced by pressure.
4. The gas hold-up ϵ_G in bubble columns increases with increasing pressure.

However, insufficient data on the influence of the operating pressure on the interfacial area in gas-liquid contactors are available.

1. INTRODUCTION

In this chapter all important studies on the influence of pressure on mass transfer phenomena in gas-liquid systems and reactors are critically reviewed. Points of agreement and conflict will be indicated and discussed in order to improve upon the understanding of the mass transfer phenomena in pressurized gas-liquid reactors. The major conclusions and points of conflict are given at the end of the chapter.

Some twenty papers have been published which deal with or touch upon the influence of the operating pressure in gas-liquid systems. They can roughly be subdivided into three groups dealing with the influence of the operating pressure on:

1. the formation and coalescence behaviour of single bubbles in gas-liquid systems.
2. the gas and liquid phase mass transfer coefficients in gas-liquid reactors.
3. the volumetric liquid phase mass transfer coefficient, the interfacial area and the gas hold-up in gas-liquid reactors.

In the next three paragraphs these groups will be discussed separately. It is important to note that in mentioning gas flow rates we will always base ourselves on the volumetric rates. This means that at constant volumetric flow rates, the mass flow rates are increased with pressure because of the increasing density. Superficial gas velocities as used by us therefore refer to the volumetric gas flow rate at the pressure in the reactor divided by the empty cross-sectional area of the reactor.

2. PRESSURE REVIEW

2.1. Formation and coalescence behaviour of single bubbles.

All studies concerning the influence of pressure on the formation of bubbles have been performed at single orifices, being connected either with a gas chamber or a capillary tube. Details of these studies like the operating pressure, the system properties and the influence of the operating pressure are summarized in Table 1.

Kling(1962) was the first one to observe that an increase in the operating pressure at equal superficial gas velocity and at a single gas inlet orifice causes a decrease in the initial bubble volume. Consequently the bubble frequency at the orifice increases. Kling(1962) suggested that the increase in energy content causes the gas to penetrate deeper into the liquid, leading to more oblong bubbles which detach easier from the orifice. This results in smaller bubbles at higher pressures.

LaNauze and Harris(1974) investigated photographically the formation of CO₂-bubbles in water at three orifices with different diameters and at pressures up to 2.1 MPa. At atmospheric pressure they observed a linear increase in the initial bubble volume with increasing gas flow rate. At higher pressures the bubble volume does not show this linear dependence anymore and an increase in the

Table 1. Studies on the formation and coalescence behaviour of single bubbles at higher pressures.

Reference	Gas system	Apparatus	Experimental method	Conditions	Remarks on the effect of a pressure increase
Kling(1962)	He/Air/N ₂ /water	orifice connected with a gas chamber $d_{or} = 1.05$ and 1.64 mm	photography	$P = 1-8$ MPa $\dot{Q}_{G,or} = 1-10$ cm ³ /s	decrease in bubble volume, increase in bubble frequency
LaNauze and Hertz(1974)	CO ₂ /water	orifice connected with a gas chamber $d_{or} = 1.6, 3.2$ and 4.8 mm	photography	$P = 1-2.1$ MPa $\dot{Q}_{G,or} = 1-30$ cm ³ /s	decrease in bubble volume at orifice of 50%, increase in bubble frequency
Sagert and Quinn(1976)	N ₂ /CO ₂ /H ₂ S/water	adjacent orifices $d_{or} = 2.0$ mm	highspeed photography	$P = 1-3.4$ MPa $\dot{Q}_{G,or} = .6$ cm ³ /s	coalescence time of N ₂ and of CO ₂ below 2.0 MPa unaffected, increase in coalescence time for CO ₂ above 2.0 MPa and for H ₂ S with a factor of 10
Bier et al(1978)	N ₂ /He/SF ₆ /water/ ethanol/refrigerant	orifice connected with a gas chamber $d_{or} = 1.6$ mm capillary tube $d_{or} = 1.6$ mm	photography	$P = 1-2.1$ MPa $\dot{Q}_{G,or} = 1-30$ cm ³ /s	decrease of bubble volume at orifice of factor 5-10, small effect with a capillary tube, agreement with Kling(1962) and LaNauze and Hertz(1974)
Kaufmann(1981)	(N ₂ /O ₂)/Ne/SO ₃ soln.	capillary tube	photography, chemically enhanced absorption	$P = 1-10$ MPa	small decrease of 25% in bubble diameter at orifice, decrease of 25% in bubble rising velocity
Idegawa et al(1986)	air/water/ethanol	capillary tube $d_{or} = 1.0$ mm	photography	$P = 1-15$ MPa $\dot{Q}_{G,or} = 1-25$ cm ³ /s	small decrease of 25% in bubble diameter at orifice, change in bubble regions, agreement with LaNauze and Hertz(1974)

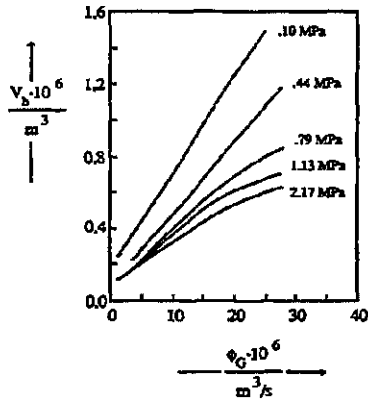


Figure 1: Bubble volume vs. the gas flow rate at different pressures; CO_2 - water, $d_{or} = 4.8$ mm (fig. 7 of LaNauze and Harris(1974)).

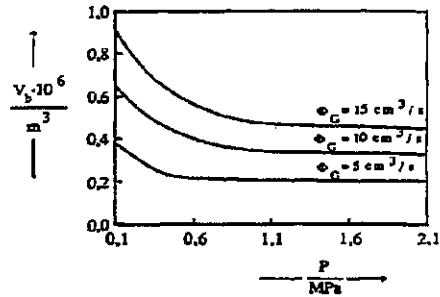


Figure 2: Bubble volume vs. the pressure at different gas flow rates; CO_2 - water, $d_{or} = 4.8$ mm (fig. 10 of LaNauze and Harris(1974)).

gas flow rate is accommodated by a simultaneous increase in the bubble volume and the bubble frequency, see Fig. 1 (figure 7 of LaNauze and Harris(1974)). With regard to the effect of the operating pressure the bubble volume decreases circa 50% with an increase from 0.1 to 1 MPa, but after that only slightly from 1 to 2.1 MPa, see Fig. 2 (figure 10 of LaNauze and Harris(1974)). This reduction in initial bubble volume results in a large degree of interaction and coalescence near the orifice at higher mass flow rates. LaNauze and Harris(1974) attributed the reduction in the size of the bubbles and the higher formation frequency to the increased contribution of the momentum of the gas to the bubble formation process as caused by higher pressures or by higher flowrates.

Experiments of Bier et al(1978) showed a rather similar dependence of the initial bubble volume on the operating pressure. Their experimental technique consisted of sparging N_2 , He or SF_6 through a single orifice into water, ethanol or a refrigerant. However, these authors find a much smaller influence of the operating pressure if the gas is sparged through a capillary tube instead of through an orifice connected with a gas chamber. This gas chamber, of course, prevents oscillations of the pressure as occur in narrow gas supply lines. Kaufmann(1981) determined the diameter of bubbles formed by injection of a N_2/O_2 mixture in a sodium sulfite solution through a capillary tube at pressures up to 10 MPa. He observed a slight decrease of 25% in the bubble diameter with a pressure increase from 0.1 to 10 MPa.

Iidogawa et al(1986) observed the bubble formation at a single orifice in air-water and air-ethanol systems and for pressures up to 15 MPa. They reported a decrease in the initial bubble diameter of 25% at a pressure increase from .1 to 15 MPa. The critical gas velocity separating the multiple bubbling and jetting regions decreased with increasing pressure. This critical gas velocity was

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correlated in an equation based on their own measurements, the results of LaNauze and Harris(1974) and on some atmospheric results as follows:

$$We_{cr, or} = 1.4 \cdot 10^4 Re_{G, or}^{-8} \quad (1)$$

Both the Weber and the Reynolds number are based on the orifice diameter and the orifice velocity, Re ranges from $4 \cdot 10^3$ up to $2.5 \cdot 10^4$. Gas velocities which result in a value below $We_{cr, or}$ and with $Re_{G, or} > 10^4$ result in the formation of single bubbles. Above the critical Weber number jetting occurs.

Sagert and Quinn(1976) measured with highspeed photography the coalescence time of two N_2 , CO_2 or H_2S bubbles in water at pressures up to 3.4 MPa. They observed no influence of the operating pressure on the coalescence time of N_2 bubbles and below 2.0 MPa no influence on the coalescence time of CO_2 bubbles. However for CO_2 the coalescence time rises from 2 - 3 ms at 2.0 MPa to 20 ms at 3.4 MPa. For H_2S bubbles the coalescence time strongly depends on the pressure and rises from around 10 ms at 0.1 MPa to 100 ms at 1.5 MPa. The authors postulated that specific interactions of the gas with the aqueous surface layers are responsible for the increase in the coalescence time.

Reference	Gas-liquid-system	d_{or}
LaNauze and Harris	CO_2 - water	1.05 mm
Kling	N_2 - water	1.6 mm
Bier et al	N_2 - water	1.6 mm

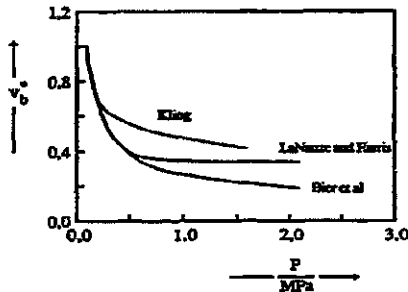


Figure 3: Normalized bubble volume vs. the pressure for an orifice connected with a gas chamber; $\dot{V}_{G, or} = 5 \text{ cm}^3/\text{s}$

Reference	Gas-liquid-system	$\dot{V}_{G, or}$	d_{or}
Kaufmann	N_2/O_2 - Na_2SO_3 sol.	$5 \text{ cm}^3/\text{s}$	
Mogawa et al	air-water	$3.2 \text{ cm}^3/\text{s}$	1 mm
Bier et al	N_2 - water	$5 \text{ cm}^3/\text{s}$	1.6 mm

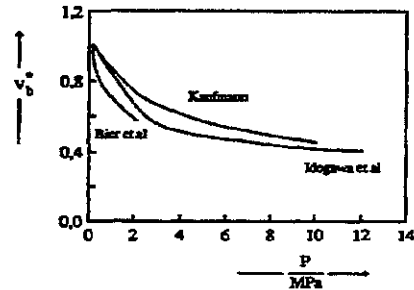


Figure 4: Normalized bubble volume vs. the pressure for a capillary tube.

All together it can be concluded that there exists a good agreement on the influence of pressure on the bubble formation at single orifices. An increase in the system pressure causes a decrease in the initial bubble size and an increase in the bubble frequency, which result in a large degree of interaction near the orifice. The effect of pressure is larger if the orifice is connected with a gas chamber instead of a capillary tube. This is shown in Figs 3 and 4 in which normalized bubble volumes of all the mentioned studies are plotted vs. pressure respectively for an orifice connected with a gas chamber and a capillary tube. The bubble volumes at a single orifice connected with a gas chamber decrease circa a factor 3 with an increase in pressure from 0.1 to 2.0 MPa. The decrease in the bubble volumes at a capillary tube is of the same order of magnitude, however, a sixfold larger increase in pressure from 0.1 to 12 MPa is needed to accomplish this. Coalescence measurements for some specific gases also indicate that the coalescence time increases at higher pressures.

2.2. Gas and liquid phase mass transfer coefficients.

In most studies the influence of the operating pressure on the gas and liquid phase mass transfer coefficients k_G and k_L is determined in stirred autoclaves with a flat surface. Vafopoulos et al(1975) are the only ones who report on liquid phase mass transfer coefficients at higher pressures in a bubble column and a mechanically agitated reactor. Details of these studies are given in Table 2.

In a stirred autoclave at pressures between 0.1 - 3 MPa Yoshida and Arakawa(1968) determined liquid phase mass transfer coefficients for the systems of oxygen in water and oxygen in an aqueous KCl solution. They observed at higher agitation rates a small decrease of k_L with pressure and at the lower agitation rate a large decrease of a factor 2 in k_L with a pressure increase from 0.5 to 2.0 MPa. They suggest that the pressure dependence of k_L is due to a change in the surface renewal rate or the degree of interfacial turbulence, which could be accounted for by the change in surface tension due to the gas pressure.

Contrarily to these results Teramoto et al(1974) using a stirred autoclave with a flat surface observed no influence on k_L of operating pressures between 0.2 - 10 MPa in absorption measurements with various gases into water, ethanol and p-xylene. This was confirmed by Albal et al(1983), who worked with O_2 -water and He-water as gas-liquid systems and at pressures between 2 - 9 MPa. For three different Fischer-Tropsch type liquids and working at pressures ranging from 1 to 5 MPa also Deimling et al(1984) mentioned that they did not observe a relation between k_L and pressure in a stirred autoclave with a flat surface. All authors explained their results by the fact that pressure does not change significantly the physical properties of a liquid and therefore does not affect the liquid phase diffusivity of the gas.

In air-water systems Vafopoulos et al(1975) determined separately both volumetric liquid phase mass transfer coefficients k_L a by physical absorption and interfacial areas a by photography. They worked in a bubble column and a mechanically agitated reactor and at pressures ranging from 0.1 to 1 MPa. From these results they calculated values for the liquid phase mass transfer coefficient

Table 2. Studies on gas and liquid phase mass transfer coefficients at higher pressures.

Reference	GM-system	Apparatus	Experimental method	Conditions	Remarks on the effect of a pressure increase
Yoshida and Araiwa (1968)	O ₂ /water/aqueous KCl solution	D = .059 m propeller stirrer	flat surface, physical absorption, increase liquid phase concentration	P = 1.3 MPa N = 75, 4.43, 6.53 rpm T = 310.5 K	small decrease k _L at higher agitation rates, large decrease k _L of factor 2 at lower agitation rate
Teramoto et al (1974)	H ₂ /He/AuCO ₂ /N ₂ /water/ethanol/p-xylole	D = .066 m turbine stirrer D ₁ = .026 m propeller stirrer D ₁ = .033 m	flat surface, physical absorption, pressure decrease	P = 2-10 MPa, T = 298 K N = 2.5, 5.0, 6.7 rpm v ₀ = 22-176 cm/s	no influence on k _L
Va (opoulos et al (1975)	(N ₂ /O ₂)/water	bubble column D _{BC} = 40 m agitated reactor D _{AR} = 44 m turbine stirrer D ₁ = .147 m porous plate (90 m), capillary tube d _{or} = 3 mm	physical absorption, liquid concentration, photography	P = 1-1 MPa N = 7.5-15.0 rpm T = 293 K H _{BC} /D _{BC} = 3.65 H _{AR} /D _{AR} = 1	no influence on k _L and a, resulting in a pressure independence of k _L
Albal et al (1983)	O ₂ /He/water	D = .102 m pitched blade stirrer D ₁ = .037 m two stirres on shaft	surface aeration, physical absorption, pressure decrease	P = 2-9 MPa N = 6.7-16.7 rpm T = 298 K, HUD = 1.34	indirect determined pressure independence of k _L
Delmink et al (1984)	CO/H ₂ /Fluor-Tropoch solutions	D = .102 m stirrer details same as above	flat surface, physical absorption, pressure decrease	P = 1-4.5 MPa T = 373-523 K HUD = 1.34	no influence on k _L
Venugog et al (1987)	(NH ₃ /NH ₄ CH ₃) / (H ₂ /He/Au/N ₂ /C ₂ H ₆) 3 M - H ₂ SO ₄ sol.	D = .081 m two turbines on shaft D ₁ = .055 m or two propellers D ₁ = .050 m	flat surface, chemically enhanced absorption, liquid concentration	P = 1-1 MPa N = 4-26.7 rpm T = 298 K	k _L inversely proportional to sqrt of total pressure, k _L ∝ P ^{-0.2}

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which appeared to be independent of pressure. Vafopoulos et al(1975) also explained the pressure independence of k_L by the fact that the liquid phase properties are not affected by pressure.

Contrarily the gas phase mass transfer coefficient k_G could be influenced indeed by pressure, because the diffusivity is inversely proportional to the total system pressure, for in ideal gases the product of the density and the diffusivity is constant, see e.g. Prausnitz et al(1978). Up till now only Versteeg et al(1987) have determined gas phase mass transfer coefficients k_G at elevated pressures between 0.1 - 1 MPa. In a stirred autoclave with a flat interface they did so by absorption of a diluted gas phase component from an inert gas phase into a liquid where an instantaneous reaction occurs between the absorbed component and a liquid-phase reactant. The gas phase mass transfer coefficient appeared to be inversely proportional to the square root of the pressure as predicted by the penetration theory, see Fig. 5.

Except for the results of Yoshida and Arakawa(1968) all studies indicate that there is no influence of the operating pressure on the liquid phase mass transfer coefficient k_L . This seems rather obvious because there is no influence of pressure on the liquid phase properties. The gas phase mass transfer coefficient k_G seems to depend on the operating pressure. The results of Versteeg et al(1987) indicate that

$$k_G = P^{-0.5} \quad (2)$$

where the value of the exponent corresponds to the predictions of the penetration mechanism.

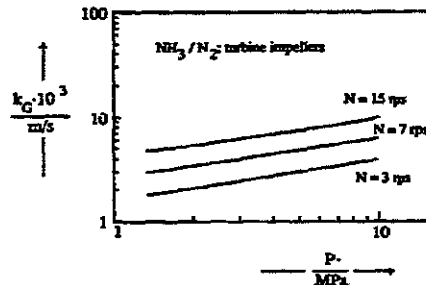


Figure 5: Gas phase mass transfer coefficient vs. pressure at different agitation rates (Versteeg et al(1987)).

Table 3. Studies on mass transfer parameters in agitated reactors at higher pressures.

Reference	Gas-system	Apparatus	Experimental method	Conditions	Remarks on the effect of a pressure increase
Ternstroem et al.(1974)	H ₂ /He/Ar/CO ₂ /N ₂ /water/ethanol/p-xylene	D = .056 m turbine stirrer D ₁ = .026 m propeller stirrer D ₁ = .033 m	surface aeration, physical absorption, pressure decrease	P = 2-10 MPa N = 23.3 rpm T = 298 K	no influence on k _L a
Vafopoulos et al.(1975)	(N ₂ O ₂)/water	D = .44 m turbine stirrer D ₁ = .147 m capillary tube d _{0T} = 3 mm	physical absorption, liquid concentration, photography	P = .1-1 MPa N = 7.5-15 rpm, T = 293 K v ₀ = 22-1.76 cm/s, HUD = 1	no influence on k _L a and k ₀
Sidhar and Potter(1980a, 1980b)	O ₂ /cyclohexane	D = .13 m turbine stirrer D ₁ = .045 m capillary tube d _{0T} = 6 mm	light transmission technique, macrometric method	P = .1-1 MPa N = 8-30 rpm, T = 297-423 K v ₀ = 1-5 cm/s, HUD = 1	k and k ₀ increase with 75%, a and k ₀ proportional to (P ₀ /P _{at}) ^{1.16}
Albal et al.(1983)	O ₂ /He/water	D = .102 m pitched blade stirrer D ₁ = .037 m two stirrer on shaft	surface aeration, physical absorption, pressure decrease	P = 2-9 MPa N = 6.7-16.7 rpm T = 298 K, initial HUD = 1.4	no influence on k _L a
Albal et al.(1984)	CO ₂ /molten paraffin wax	D = .102 m stirrer details same as above	surface aeration, physical absorption, pressure decrease	P = 1-3 MPa N = 13.3, 16.7 rpm T = 348, 423 K, initial HUD = 1.4	no influence on k _L a
Detmeling et al.(1984)	CO ₂ /H ₂ /Fischer-Tropsch solutions	D = .102 m stirrer details same as above	surface aeration, physical absorption, pressure decrease	P = 1-4.5 MPa N = 13.3, 16.7, 18.3 rpm T = 373-523 K, initial HUD = 1.34	increase k _L a of a factor 1.5 for the light fraction and a factor 4 for the heavy fraction of a Fischer-Tropsch liquid
Leszkowicz et al.(1984)	H ₂ /CO/CO ₂ /N ₂ /molten wax	D = .08 m turbine impeller D ₁ = .048 m	surface aeration, physical absorption, pressure decrease	P = .5-6 MPa N > 11.7 rpm T = 473-493 K	no influence on k _L a
Karnoolkar et al.(1986)	CO/H ₂ /Fischer-Tropsch solutions	D = .127 m turbine impeller D ₁ = .0635 m	surface aeration, physical absorption, pressure decrease	P = 1-4 MPa N = 11.7, 16.7, 20 rpm T = 423-498 K, HUD = 1.5	increase k _L a of a factor 2 at all agitation rates

2.3. Volumetric liquid phase mass transfer coefficients, interfacial areas and gas hold-ups.

Most studies in pressurized gas-liquid systems concern the influence of pressure on the overall mass transfer parameters $k_L a$, a , and the gas hold-up ϵ_G in agitated reactors and bubble columns. They are discussed in the next two paragraphs and the details are summarized in Tables 3 and 4 respectively. It will be shown that there is no general agreement among the several authors on the effect of pressure on the mass transfer parameters in these reactors.

2.3.1. Agitated reactors.

Four recent studies on mass transfer coefficients in stirred autoclaves for the gases H_2 and CO and Fischer-Tropsch type liquids are an example of the contradictory results concerning the effect of pressure. Albal et al(1984) determined volumetric liquid phase mass transfer coefficients $k_L a$ for H_2 and CO in molten paraffin wax and at pressures ranging from 1 - 3.5 MPa. They worked under conditions of surface aeration and observed that $k_L a$ was independent of the operating pressure. Deimling et al(1984) used the same installation and determined $k_L a$ for H_2 and CO in three Fischer-Tropsch type liquids and at pressures ranging from 1 to 4.5 MPa. At an increase in pressure from 1 to 4 MPa for both gases they observed a strong pressure effect resulting in an increase of $k_L a$ by circa a factor 1.5 for the light fraction and a maximum of 4 for the heavy fraction. According to these authors this is mainly due to the effect of pressure on the interfacial area a . They suggest that a increases with pressure because of the change in surface tension, which is around 40% for the pressure range and liquids used. This results in the formation of smaller entrained bubbles. Addition of inert solids did not change this strong pressure effect. Ledakowicz et al(1984), using the same experimental technique as mentioned above, reported on $k_L a$ data for H_2 , CO, N_2 and CO_2 in a molten wax under Fischer-Tropsch conditions and pressures between 0.1 - 6 MPa. They did not observe any effect of pressure. Contrarily at pressures between 1 - 4 MPa Karandikar et al(1986) again found a pronounced influence of pressure on $k_L a$ for H_2 and CO in a medium fraction of Fischer-Tropsch type liquid. At all agitation rates their data show an increase of $k_L a$ with a factor of 2 at a pressure increase from 1 to 4 MPa.

All studies are summarized in Fig. 6 in which a normalized volumetric liquid phase mass transfer coefficient is plotted vs. pressure. Karandikar et al(1986) did not give an explanation for the pressure effect, but from these four studies in which the same experimental method and gas-liquid systems are used, it is evident that - even if there is any effect of pressure on $k_L a$ - its magnitude is probably dependent on the specific surface properties of the liquid phase.

The same experimental technique has also been used by Albal et al(1983) and by Teramoto et al(1974) to determine $k_L a$ data for H_2 , O_2 and N_2 in water. They worked at pressures from 0.2 to 10 MPa and both groups did not find any influence of pressure on $k_L a$.

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Contradictory results regarding the influence of pressure can also be found in sparged mechanically agitated reactors. Working with an air-water system and at pressures between 0.1 - 1 MPa Vafopoulos et al(1975) did not find any influence of pressure on $k_L a$, α and ϵ_G in a mechanically agitated reactor. Within the same pressure range Sridhar and Potter(1980a, 1980b) observed an increase of as much as 75% of the interfacial area in a mechanically agitated reactor with the system O_2 -cyclohexane. Sridhar and Potter(1980a, 1980b) attributed this increase of the mass transfer parameters to the increase with pressure of the kinetic energy content of the inlet gas flow. To correlate their results they modified the equations of Calderbank(1958) by multiplying them by a factor $(E_T/P_G)(\rho_G/\rho_{air})^{0.16}$, where the first factor represents the ratio of the total (kinetic and mechanical) energy supplied to the dispersion and the power input by agitation only. A second correction factor for the gas density had to be applied, so it was not possible to account for the influence of pressure solely by the increase in the kinetic energy content of the gas flow.

Re. source	Liquid	N
Albal et al	paraffin wax	6.7-16.7 rpm
Lodakowicz et al	paraffin wax	> 11.7 rpm
Deimling et al	Fischer-Tropsch type liquids	13.3 rpm
Karamoliar et al	Fischer-Tropsch solution	16.7 rpm

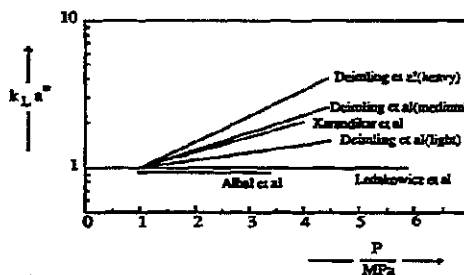


Figure 6: Normalized volumetric liquid phase mass transfer coefficient vs. the pressure in surface aerated agitated reactors.

2.3.2. Bubble columns.

That not only the increase in the kinetic energy content is responsible for the increase in gas hold-up with pressure has been confirmed for a bubble column by Pijls et al(1985). They found a twofold increase in gas hold-up for a pressure increase from 0.1 to 2 MPa. They used a N_2 -water system and different sparger rings and concluded that neither an increase in the kinetic energy nor in the momentum of the gas flow can account for the increase in the gas hold-up. They also observed a smaller effect of pressure with a sintered glass plate than with the sparger rings. Pijls et al(1985)

Table 4. Studies on mass transfer parameters in bubble columns at higher pressures.

Reference	Gas-liquid system	Apparatus	Experimental method	Conditions	Remarks on the effect of a pressure increase
Köhler et al (1961)	Air/water	D = .192 m porous plate (50-100 μ m)	height difference, photography	P = 1-1.6 MPa Initial H/D = 4.7 V _G = 1-3.5 cm/s	no influence on d_b and ϵ_G
Penkov and Koiokhtsev (1965)	air/water	column .1 ^o .1 m perforated plate (25 holes of 3 mm)	X-ray attenuation	P = 1-2.5 MPa V _G = .01-.4 cm/s T = 293 K, H/D = 7	ϵ_G proportional to $(\rho_G / \rho_{AIR})^{.22}$
Václavík et al (1975)	(N ₂ /O ₂)/water	D = .40 m porous plate (90 μ m), capillary tube d _{or} = 3 mm	physical absorption, liquid concentration, photography	P = 1-1 MPa T = 293 K, H/D = 3.65 V _G = .67-2.65 cm/s	no influence on k_L , a , and ϵ_G
Deckwer et al (1980)	N ₂ /N ₂ O ₂ /melted paraffin wax	D = .041 or .10 m porous plate (75 μ m)	height difference	P = 4-11 MPa T = 523 K, H/D = 6-10 V _G = .5-3.4 cm/s	no influence on ϵ_G
Tanny et al (1984, 1984b)	N ₂ /ethylene	D = .61 m bubble caps	height difference	P = .121-.62 MPa T = 298 K, H/D = 7 V _G = 3-16 cm/s	increase in ϵ_G of a factor 2, change in hydrodynamics
Iogawa et al (1985a)	air/water	D = .05 m, porous plates (2, 100 μ m) capillary tubes (1, 3 and 5 mm) perforated plate (19 holes of 1 mm)	electric resistivity probe (two points)	P = 1-1.5 MPa T = 288-293 K, H/D = 16.6 V _G = 5-5 cm/s	ϵ_G and bubble frequency increase and d_b decreases, above 5 MPa uniform bubble size
Iogawa et al (1985b)	H ₂ /He/air/water/methanol/ethanol/acetic acid, etc. sol.	D = .05 m perforated plate (19 holes of 1 mm)	electric resistivity probe (two points)	P = 1-5 MPa T = 284-293 K, H/D = 16.6 V _G = 5-5 cm/s	ϵ_G and bubble frequency increase and d_b decreases, empirical correlations given in text
Pijla et al (1985)	N ₂ /water	D = .158 m, different gas sparger rings and stainless glass plate (200-500 μ m)	overflow tube	P = 1-2 MPa T = 293 K, H/D = 9.4 V _G = 4-15 cm/s	ϵ_G increases with a factor of 2 if sparger rings are used, increase smaller if altered plate is used
Teunissen et al (1986)	N ₂ /water	D = .158 m, gas distributors same as above	five points resistivity probe	P = 1-2.0 MPa T = 293 K, H/D = 7 V _G = 2-20 cm/s	ϵ_G increases, d_b decreases, consequence not affected by pressure, change in hydrodynamics

explain their results qualitatively by a simultaneous decrease of the initial diameter and of the coalescence rate of the bubbles with an increase of pressure. Teurlings et al(1986) determined bubble diameters in the same experimental system with a multipoint resistivity probe. Again they found a pronounced effect of pressure on the gas hold-up. This influence was smaller for the sintered glass plate as gas distributor. They observed with increasing pressure a decrease of the average bubble diameter in the column. Bubble size distributions were measured at different heights in the bubble column and at different pressures: it could be concluded that the coalescence of bubbles was not influenced by pressure. Teurlings et al(1986) also observed that with increasing pressure the fraction of fast moving bubbles becomes smaller. According to them this indicates a change in the hydrodynamics: at higher pressures the transition from the bubbling regime towards the churn-turbulent regime, see Shah et al(1982), occurs at higher gas velocities.

Tarmy et al(1984a, 1984b) reported on gas hold-up measurements in a bubble column with N_2 -heptane as the gas-liquid system and bubble caps as gas distributors. They varied the pressures between 0.12 - 0.62 MPa and found a strong influence of pressure. The gas hold-ups at 0.62 MPa were about twice those at the pressure of 0.12 MPa. Also Tarmy et al(1984a, 1984b) calculated that a change in flow regimes occurs: at higher pressures the transition from the uniform bubbling regime towards the churn-turbulent regime occurs at higher gas velocities and gas hold-ups, see Shah et al(1982). According to them the fact that the bubbling regime persists at higher gas hold-ups indicates that at higher pressures very small, non-coalescing bubbles are present in the system.

With an electric resistivity probe Idogawa et al(1985a) studied the behaviour of bubbles in the air-water system in a bubble column. They used different gas distributors and worked at pressures ranging from 0.1 to 15 MPa. With an increase in pressure the gas hold-up and bubble frequency increased, while the bubble size decreased. Above 5 MPa the bubble size distributions become narrow and the size of the bubbles almost uniform. The effect of pressure was much smaller when porous plates were used as gas distributors instead of single orifices or a perforated plate. In a second study Idogawa et al(1985b) extended their research to other systems, consisting of H_2 , He or air as gases and of water, methanol, ethanol, acetone or aqueous solutions of alcohols as liquids. In these experiments the pressure ranged from 0.1 to 5 MPa and as gas distributor a perforated plate with 19 holes of 1 mm diameter was used. The following correlations were obtained for the gas hold-up and the average bubble diameter:

$$\frac{\epsilon_G}{(1 - \epsilon_G)} = 1.44 v_G^{0.58} \rho_G^{0.12} \sigma_L^{-0.16} \exp(-P) \quad (3)$$

$$d_b = 3.10 \cdot 10^{-3} \rho_G^{-0.028} \sigma_L^{0.088} \exp(-P) \quad (4)$$

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In these expressions SI units have to be used, except for the pressure P and the surface tension σ_L , which have to be expressed in MPa and mN/m respectively. Their experimental range was $v_G = 0.5 \cdot 10^{-2} - 5 \cdot 10^{-2}$ m/s, $\rho_G = 0.084 - 120.8$ kg/m³, $\sigma_L = 22.6 - 72.1$ mN/m and $P = 0.1 - 5$ MPa. For air-water at a constant superficial gas velocity of 1 cm/s these correlations predict an increase in the gas hold-up with a factor 2.7 and a decrease in the average bubble diameter with a factor 0.6, when the pressure is increased from 0.1 up to 5 MPa: this results in an increase with a factor 4.5 in the interfacial area.

Petuhkov and Kolokol'tsev(1965) studied liquid entrainment in a bubble column with air-water as the gas-liquid system and at pressures from 0.1 to 2.5 MPa. At pressures of 0.1, 0.3 and 2.5 MPa they also determined the gas hold-ups and found that the gas hold-up was proportional with $(\rho_G / \rho_L)^{0.22}$.

All these six studies indicate that there is an effect of pressure on the gas hold-up and bubble diameters in a bubble column, see Fig. 7. In contradiction with these studies Kölbl et al(1961), Vafopoulos et al(1975) and Deckwer et al(1980) did not find any influence of pressure on the gas hold-up and bubble diameters in two and three phase bubble columns.

Reference	GL-system	gas distributor	v_G
Pijls / Teurlings et al	N ₂ / water	sparger ring 66°2.75 mm	10 cm/s
" "	"	sintered glass plate(200-500 μ m)	10 cm/s
Tanay et al	N ₂ / heptane	bubble caps	10 cm/s
Idogawa et al	air / water	perforated plate: 19°1 mm	5 cm/s
"	"	porous plate(100 μ m)	3 cm/s
Petuhkov and Kolokol'tsev	air / water	perforated plate: 125°3 mm	.01-4 m/s

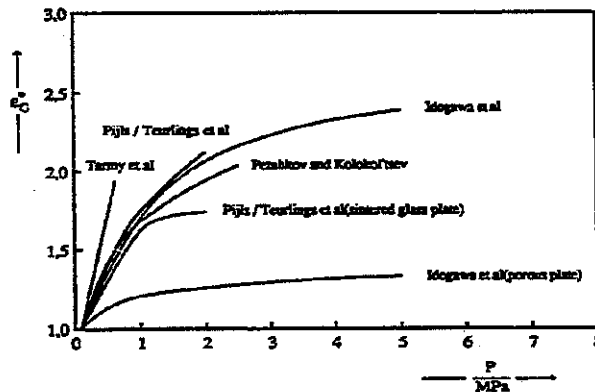


Figure 7: Normalized holdup vs. the pressure in bubble columns.

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Kölbel et al(1961) worked at pressures between 0.1 - 1.6 MPa in a bubble column with air-water as gas-liquid system and a porous plate as gas distributor. They determined bubble diameters by photography and could not find any influence of pressure. They also measured gas hold-ups by the height difference method and found them to be independent of the operating pressure. Vafopoulos et al(1975), who used the same experimental technique, also reported bubble diameters and gas hold-ups to be independent of pressure in a bubble column. They worked also with air-water as gas-liquid system and a porous plate or a capillary tube as gas distributor at pressures from 0.1 to 1 MPa. Besides bubble diameters and gas hold-ups Vafopoulos et al(1975) determined by physical absorption volumetric liquid phase mass transfer coefficients $k_L a$ and found them to be independent of pressure.

Deckwer et al(1980) studied the influence of pressure on the gas hold-up at pressures from 0.4 to 1.1 MPa in two bubble columns with a porous plate as gas distributor and the three-phase system of N_2 - Al_2O_3 -molten paraffin wax. In agreement with Kölbel et al(1961) and Vafopoulos et al(1975) they did not find any influence of the operating pressure on the gas hold-up.

All together there seems to be a lot of disagreement between the several authors on the influence of pressure on the overall mass transfer parameters $k_L a$, a and the gas hold-up ϵ_G in agitated reactors and bubble columns. However when the operating conditions and experimental systems are considered more closely some specific remarks can be made as will be done in the next paragraph.

3. DISCUSSION

All studies on *the bubble formation at a single orifice* in a pressurized gas-liquid system indicate a decrease in the initial bubble size when the pressure increases. This decrease is larger when the orifice is connected with a gas chamber instead of using only a capillary tube. Modelling of the experimental results was only partly successful. LaNauze and Harris(1974) modelled their experiments at higher pressures with a modified version of the bubble growth model of Davidson and Schiller(1960). They extended this model to allow for the rate of change of gas momentum and for varying gas chamber pressures. Their model showed good agreement with their experimental results at lower flow rates, but discrepancies occur at higher flowrates, because the theory deals with single bubbles only and assumes no interaction of bubbles. For an orifice connected with a gas chamber as well as their results for a capillary tube Bier et al(1978) compared their results with the semi-empirical diagrams for bubble formation at a single orifice as given by Mersmann(1977). These diagrams are based on the two step bubble formation model of Kumar and Kolloor(1970), which assumes a constant gas flow rate during the bubble formation process. An excellent agreement was found between the experimental results with a capillary tube and the prediction by the diagrams, while for an orifice connected with a gas chamber the results showed considerable deviations. Bier et al(1978) argued that this probably is caused by the fact that the gas flow

rate is irregular during the bubble formation at an orifice connected with a gas chamber, while it is approximately constant for the capillary tube. Finally they correlated their results successfully with an empirical correlation, but unfortunately this correlation is specific for their experimental configuration and conditions.

Therefore at this moment it seems not possible to predict the initial bubble volume at a single orifice in pressurized gas-liquid systems. However, the qualitative results can be used for a better understanding of the influence of pressure in gas-liquid reactors on the overall mass transfer parameters $k_L a$, α and the gas hold-up ϵ_G . Besides this, one may wonder whether correlations for the initial bubble diameter are useful to the designer of pressurized gas-liquid reactors, because considerable coalescence is known to take place in the vicinity of the gas sparger.

The influence of pressure on the gas and liquid phase mass transfer coefficients k_G and k_L can be separated in an effect of pressure on the hydrodynamics of the phase and an effect of pressure on the physical properties of the phase under consideration. Important liquid phase properties like the viscosity, the specific gravity and the diffusivity of the transferred component in the liquid phase are only affected considerably at extremely high pressures. Contrarily the gas phase density and the diffusivity in the gas phase are affected by pressure indeed and approximately proportional to pressure. As shown by Versteeg et al(1987), see Fig. 5, this results in a considerable influence of pressure on the gas phase mass transfer coefficient k_G .

The changes with pressure in the bubbling regimes at a single orifice and in the flow regimes in a bubble column as reported respectively by Idogawa et al(1986) and Tammy et al(1984a, 1984b) indicate the possibility of an effect of pressure on the hydrodynamics in gas-liquid reactors. This most certainly will affect the characteristic mass transfer parameter (film thickness or surface renewal time) of the prevailing mass transfer model or even the applicability of the mass transfer model itself. Changes with pressure in the gas hold-up and bubble diameters may also influence the mass transfer coefficients. Furthermore Massoudi and King(1974) and Sagert and Quinn(1978) reported an influence of pressure on the surface tension and also on the apparent viscosity of the surface of different gas-liquid systems, which may affect the hydrodynamics of the phases. With this in mind, we should be very careful in stating that the liquid phase mass transfer coefficient k_L is independent of pressure as long as we are not absolutely sure that the hydrodynamics of the gas-liquid system are not affected by the pressure.

The results in *surface aerated agitated reactors* for the four studies under Fischer-Tropsch conditions and the two studies with water show no similar effect of pressure on the volumetric liquid phase mass transfer coefficient $k_L a$. However, there seems to be a specific relation between the effect of pressure and the liquid phase used. It is possible to distinguish between the studies with Fischer-Tropsch type liquids (Deimling et al(1974) and Karandikar et al(1986)) and with water (Teramoto et al(1974) and Albal et al(1983)) and molten waxes (Albal et al(1984) and

Ledakowicz et al(1984)) respectively. The studies with the Fischer-Tropsch type liquids show a pronounced effect of pressure, which is even dependent on the composition of the liquid, see Fig. 6 and Deimling et al(1984). Both the studies in water and with molten waxes show no influence of pressure at all. In our view this indicates that specific gas-liquid interaction parameters like the surface tension and the foaming characteristics of the liquid could have a strong influence on the observed pressure effect. Whether these specific gas-liquid interactions are also the reason for the contradictory results found in sparged agitated reactors by Vafopoulos et al(1975) and Sridhar and Potter(1980a, 1980b), who used water and cyclohexane respectively, cannot be concluded.

A critical review of all studies on the influence of the operating pressure on *the overall mass transfer parameters $k_L a$, a and the gas hold-up ϵ_G in bubble columns* leads to the following three remarks:

1. All three studies, which claimed that no influence of pressure exists, used a porous plate (< 100 μm) as gas distributor. The other studies used different gas distributors and in case they too used porous plates, they observe a much smaller effect of pressure, see Fig.7 (sintered glass plate(200-500 μm) as discussed by Pijls et al(1985) and Teurlings et al(1986); porous plates(2, 100 μm) by Idogawa et al(1985a)).
2. It is not possible to make a distinction between the studies in bubble columns on the basis of the liquids used, as was the case with the surface aerated agitated reactors. K ilbel et al(1961) and Vafopoulos et al(1975) used water as liquid and did not find an effect of pressure. On the contrary Petukov and Kolokol'tsev(1965), Idogawa et al(1985a, 1985b), Pijls et al(1985) and Teurlings et al(1986) all found a pronounced effect of pressure in their water containing systems. The same findings hold for organic liquids, where Tarmy et al(1984a, 1984b) and Idogawa et al(1985b) found a pronounced pressure influence, while Deckwer et al(1980) observed no effect at all.
3. In all three studies, which claimed that no influence of pressure exists, low superficial gas velocities between 0.1 - 3.5 cm/s were applied. In most studies, in which an influence of pressure was found, higher gas velocities between 2 - 20 cm/s were used. The studies of Idogawa et al(1985a, 1985b), who found a pressure effect working with rather low superficial gas velocities between 0.5 - 5 cm/s, are an exception.

Bubbles formed at a porous plate have a rather broad size distribution and show much interaction with each other on the surface of the porous plate. This could easily lead to coalescence of the bubbles and so disguise an effect of pressure on the bubble formation process at a porous plate. This could be the reason that most studies with porous plates show no effect of pressure on $k_L a$, a and ϵ_G . Another possibility is that an effect of pressure may occur only at higher superficial gas velocities.

The change in flow regimes with an increase in pressure as reported by Tarmy et al(1984a, 1984b) and Teurlings et al(1986) can be explained by the formation of smaller bubbles at higher

pressures. These bubbles have lower rising velocities leading to an increase in the gas hold-up. It is interesting to note that this may result in the operation of a bubble column still in the bubbling regime at high gas hold-ups and at higher pressures. In the work of Tarmy et al(1984a, 1984b) it can be seen that at a gas hold-up of $\epsilon_G = 0.3$ and at $P = 0.62$ MPa the bubble column still operates in the bubbling regime, while at atmospheric pressure the change from the bubbling regime towards the churn-turbulent regime normally occurs at gas hold-ups between 0.10 - 0.15, see Shah et al(1982). The combination of decreasing bubble diameters and increasing gas hold-ups can result in a spectacular increase in the interfacial areas. This was already shown with the correlations of Idogawa et al(1985b) in Eqs (3) and (4), which showed an increase of a factor 4.5 in the interfacial area with an increase in pressure from 0.1 to 5 MPa for water and at $v_G = 1$ cm/s.

4. CONCLUSIONS

We may draw the following conclusions after reviewing the papers mentioned:

1. The initial bubble size at a single orifice decreases with increasing pressure. The effect is larger if an orifice connected with a gas chamber is used instead of a capillary tube.
2. The critical gas velocity separating the multiple bubbling and jetting regimes at a single orifice decreases with increasing pressure.
3. The gas phase mass transfer coefficient k_G is inversely proportional with pressure to the power 0.5. This exponent is in accordance with the penetration theory.
4. The liquid phase mass transfer coefficient k_L is not influenced by pressure provided the liquid phase hydrodynamics are unaffected by pressure.
5. The influence of pressure on the volumetric liquid phase mass transfer coefficient $k_L a$ in surface aerated agitated reactors seems to depend on specific gas-liquid interaction parameters like the surface tension and the foaming characteristics.
6. The gas hold-up ϵ_G in bubble columns increases with increasing pressure. This pressure influence is absent if a porous plate is used as gas distributor.
7. In bubble columns at higher pressures the transition from the uniform bubbling regime towards the churn-turbulent regime occurs at higher superficial gas velocities and at higher gas hold-ups than at atmospheric pressure.

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NOTATION

a	specific interfacial area, m^2/m^3 dispersion
d	diameter, m
D	reactor diameter, m
D_i	impeller diameter, m
E_T	total energy input into the dispersion, W
H	dispersion height, m
k	mass transfer coefficient, m/s
n	exponent
N	agitation rate, s^{-1}
P	pressure, Pa
P_G	gassed power input, W
$Re_{G, or}$	orifice Reynolds number, $(v_{G, or} d_{or} / \nu_G)$
T	temperature, K
v	superficial velocity based on the empty cross-sectional area of the vessel, m/s
V	volume, m^3
We_{or}	orifice Weber number, $(\rho_L v_{G, or} d_{or} / \sigma)$

Greek symbols

ϵ	hold-up
ϕ	flow rate, m^3/s
ν	kinematic viscosity, m^2/s
ρ	density, kg/m^3
σ	surface tension, N/m

Subscripts

air	air
AR	agitated reactor
b	bubble
BC	bubble column
cr	critical
G	gas
L	liquid
or	orifice

Superscripts

*	normalized to atmospheric conditions
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