Research Article

Effect of Gas Permeability and Solubility on Foam

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We perform a study on the influence of gas permeability and solubility on the drainage and stability of foam stabilized with an anionic surfactant. Our study compares the foam stability for four pure gases and two gas mixtures while previous works only compared two pure gases. Drainage and foam-volume-decay rates are calculated from the experimental data and analysed. We find good agreement with existing theory as the foam stability is strongly influenced by the properties of the gas phase, in particular its solubility in the aqueous phase (measured by Henry's solubility constant, k_H) and permeability (measured by foam-film permeability coefficient, K). The foam volume decreases considerably with increasing K. Moreover, we observe that foams are more stable when a less soluble gas is added to a more soluble gas. Our analysis confirms theories linking drainage, stability, and coarsening rate. Finally, we introduce a new formulation for the foaming index that considers gas solubility and permeability.

1. Introduction

Aqueous foams are dispersions of a gas in a surfactant solution (containing water, surfactant, and possibly electrolyte or particles) [1–3]. Similar to the liquid phase, the gas phase might consist of more than a single component. For instance, when foam is applied in petroleum industry for improving the oil production, the gases are often mixtures of a number of gases. Another example includes direct utilization of the flue gas (mixture of N_2, CO_2, and NO_x's) in several applications, which aims at reducing costs of separation of CO_2 from flue gas.

Foam can be characterized by physicochemical properties of its constitutive components, such as bubble shape and size, liquid fraction, and film thickness [1, 2, 4]. The properties of both phases (and the components in the phases) control the dynamics of foam behaviour and eventually affect foam longevity. While many studies have shown the impact of the components of the aqueous phase on foam stability [1, 4–9], the effect of type and composition of the gaseous phase has received less attention.

Foam stability is controlled by three main factors: drainage, coarsening, and bubble coalescence. Coarsening refers to the growth of the average bubble size. Two processes are responsible for the changes in degree of dispersion of gas bubbles in foam: (i) the diffusion of gas through the lamellae and (ii) collapse of liquid lamellae and subsequent coalescence of contiguous gas bubbles. Pressure difference between bubbles of unequal size induces gas transfer from small to larger bubbles [10–13].

The ability of gas to permeate through a foam film is measured by a constant referred to as the "permeability coefficient, K," which depends on the properties of the foam components, for example, type of surfactant, concentration of surfactant and electrolyte in the foaming solution, temperature, and solubility of the gaseous phase in the aqueous phase [11–23]. It appears from experimental data that K is directly proportional to the product of the molecular diffusion coefficient, D, of the gas in the aqueous phase and Henry's solubility constant (k_H), that is, $K \propto Dk_H$. Indeed, on a log-log plot K varies linearly with Dk_H [23]. In some literature the coarsening rate of foam has been quantified

TABLE 1: Properties of the gases used in the experiments. K is the single foam film permeability to gas, k_H is Henry's solubility constant, D is the molecular diffusion coefficient, and t_c is critical time of coarsening.

Gas type	K[1] (m/s)	k_H[2] (—)	μ[3] (10^{-3} cP)	D[3] (10^5 cm^2/s)	$D \times k_H$ (10^5 cm^2/s)	t_c[7] (s)
CO_2	7.85	0.831	14.80	1.90	1.580	5
He	0.398	0.0093	19.41	6.8	0.063	1440
Flue gas	0.163[4]	0.177	16.78[6]	1.98	0.350	179
N_2	0.131	0.0132	17.40	2.0	0.026	118
CH_4	0.30	0.032	10.87	1.90	0.061	308
50% CH_4-50% CO_2	0.5767[4]	0.416	13.32[6]	1.97[5]	0.820	65

[1] Data taken from Princen et al., 1976 [12], and Princen and Mason, 1965 [13].
[2] Data taken from Sander, 1999 [46].
[3] Data taken from CRC handbook of chemistry and physics, 1980 [47].
[4] Calculated using (1).
[5] Perry's chemical engineering handbook, chapter 2 [48].
[6] Calculated by (7) of Norman et al., 1954 [49].
[7] Calculated from (3) of Hilgenfeldt et al., 2001 [32].

using an "*effective*" diffusion coefficient, D_{eff}, which depends on the ideal gas molar volume, Henry's law constant, and the gas permeability through the film [24].

For a mixture of gases, the behavior of each component gas needs to be accounted for. Princen et al. [12, 13] showed that the permeability coefficient of a film to a mixture of gases can be calculated using the following equation:

$$K_G = \left(\sum_{i=1}^{n} \frac{x_i}{K_i} \right)^{-1}, \qquad (1)$$

where n is the number of components and x_i and K_i are the mole fraction and permeability of the film to component i, respectively. Princen and Mason [13] argued that the gases diffuse out of the bubble at nearly the same ratio as their respective mole fractions inside the bubble, so that the mole fractions subsequently change very slowly. An important implication of (1) is that the diffusion of a fast-permeating gas can be slowed down by admixing it with a slow-permeating gas, which leads to more stability of the foam [25].

The transfer of gas through lamellae is governed by the thermodynamics of the single foam films; that is, the interactions of the two adsorbed monolayers result in slower coarsening rates than the Fickian diffusion [26–29]. Films also rupture when the liquid fraction is too small for transient films to be formed [27, 29–31].

The coarsening depends on liquid fraction, bubble size, the chemistry of the liquid, and the gas type [14–23]. Note that drainage, coarsening, and coalescence are coupled phenomena. During foam coarsening, gravity-driven drainage redistributes the liquid in the foam. After a certain time, drainage rate decreases and coarsening becomes significant. This time is referred to as the critical time of coarsening, t_c [32]. The t_c of the gases used in this work is listed in Table 1. Saint-Jalmes et al. saturated the N_2 gas with SF_6 or C_2F_6 to suppress the coarsening and study the modification of the drainage [24, 33–36]. It has been observed that the gas with higher solubility leads to a foam with higher drainage rate [15, 24]. Similar behavior has been observed in emulsions,

where Ostwald ripening is limited by addition of an insoluble oil [37]. Gas solubility also affects foam stability in porous media [23, 25, 38–43], although the underlying mechanisms are not as straightforward as the foam in bulk [39].

Previous works have demonstrated that strong coarsening accelerates the drainage and shortens the foam lifetime [24, 32, 44, 45]. These studies have compared gases with large difference in their solubilities such as CO_2 and C_2F_6. Furthermore, the effects of gas solubility and permeability on the foamability have not yet been quantified.

Our objective is therefore to experimentally investigate the effect of gas type and composition on foam generation, drainage, coarsening, and coalescence. Foam was created by gas sparging and foam volume and liquid fraction was continuously monitored. Experiments were conducted with gases of different solubility and permeability in water and with mixtures of gases. The foams were characterized by their foaming time, volume, drainage rate, and the permeability of their single films to the respective gas. The structure of the paper is as follows. First we describe the experimental setup and the material used in the experiments. Afterwards, we examine foaming and the stability of foams and relate foam physics to the properties of the gases. Finally, we end the paper with concluding remarks.

2. Experiments

2.1. Material. The surfactant used was (C_{14}–C_{16})-alpha-olefin sulfonate, AOS (Stepan Company, USA). This surfactant is anionic with industrial purity and was used as received without any further purification. The molecular weight of the surfactant is $M_w \approx 315$. Properties of the solution and single foam films stabilized by this surfactant can be found in [50]. Sodium chloride (NaCl) was used as electrolyte. The surfactant solution used in all experiments contained 2 wt% AOS and 3 wt% NaCl.

Surface tension was measured only in air and was not measured in the presence of the different gases used in foam generation.

All the gases (CO_2, CH_4, N_2, and He) were with purities of above 99%. For the gas mixtures, the first was a synthetic flue gas made from N_2 and CO_2 (in an 80%/20% molar fraction mix), and the second was a 50%/50% mixture of CO_2 and CH_4. The main properties of the gases, and gas mixtures, are summarized in Table 1. The film permeability coefficient, K, of the gas mixtures was calculated using (1). Henry's solubility constant, k_H, and the molecular diffusion coefficient, D, of the gas mixtures were assumed to be a linear function of the molar fraction of the pure gases. All the experiments were performed at atmospheric pressure and room temperature ($\sim 20°C$).

2.2. Foaming Setup.

This study was carried out using the *Foamscan* apparatus (manufactured by Teclis), an experimental setup commonly used to study foam drainage and stability [5, 51, 52]. The foam was generated in a glass column (with an inner diameter of 3 cm) by sparging gas though a glass frit (with a thickness of 3 mm and pore sizes in the range 40–100 μm) at the bottom of 50 mL of surfactant solution. During foam generation the gas was injected at a fixed rate of 10 mL/min, and the system was set to stop injection automatically after the foam volume in the column reached 100 mL.

A pair of electrodes at the bottom of the column measured the volume of the free liquid. A camera measured the total height of the foam top. The foam volume was calculated by subtracting the liquid volume from the total volume. Additionally, a conductimetry technique [53] was used to measure the liquid fraction ε, with two pairs of electrodes positioned on opposing sides of the column to measure the liquid fractions during each experiment.

To check the repeatability of the experiments, each experiment was run twice. The difference between repetitions was of the order of a small percent.

2.3. Foaming Index.

The foaming properties of gases with different solubility in water were tested using the same surfactant solution and foaming procedure for each gas. The foamability (ability to create the foam) was quantified by the foaming index (FI), defined as the ratio of the foaming rate, $r_f(t)$ (the volume of foam, $V_f(t)$, created per unit time, i.e., $r_f(t) \equiv dV_f(t)/dt$), and the rate at which gas was injected $r_g(t)$:

$$\text{FI} \equiv \frac{r_f(t)}{r_g(t)} \equiv \frac{V_f(t)}{V_g(t)}. \tag{2}$$

The FI is similar to the coefficient Ω introduced by Bikerman in 1938 [54] to describe foam behavior during steady-state experiments. In contrast the FI can be used to describe transient, or foaming phase, experiments. Hrma [55] and Capek [37] have disputed the consistency of FI due to its dependency on the gas rate. However in this study all the experiments were conducted with the same gas injection rate and therefore FI is a good measure of foamability of the gases. The FI can be larger than unity when small amounts of water are entrained within the bubbles. In this case foaming is assumed to be very efficient. On the contrary, FI below
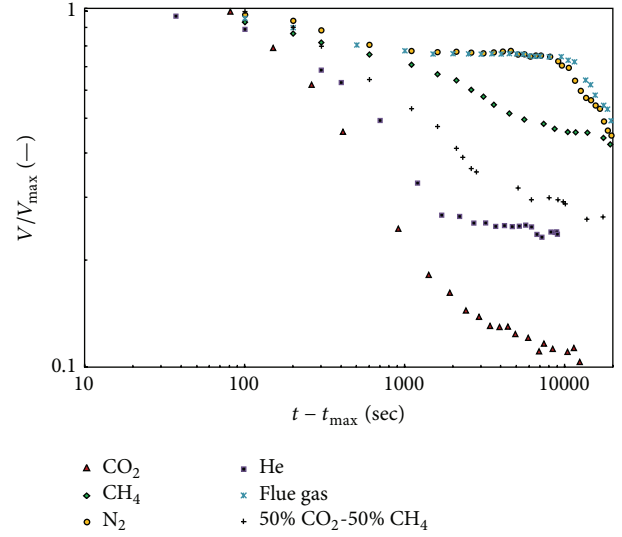

FIGURE 1: Evolution of foam with time for different gases. To eliminate the generation stage, foam volume has been plotted versus ($t − t_{\max}$), where $t_{\max}$ is the time at which foam volume reaches its maximum value in the experiment.

unity indicates collapse of foam and implies lower foaming efficiency. In this case a large volume of gas is required to create a certain foam volume.

3. Results and Discussion

3.1. Foam Generation and Destruction.

An important feature of foam is its irreversible evolution with time to achieve minimum free energy through minimization of its surface area [4]. For all our experiments, the maximum volume of the foam was set to be 100 mL. We assign $t_{\max}$ to be the time at which the foam volume reached this maximum value for the constant gas injection rate used. Once the foam volume reached its maximum value the gas injection stopped.

The general characteristics of the experiments for all gases are largely similar. Foam volume increases with injection of gas, more strictly after the foaming solution is saturated with the injected gas. Once the foam volume reaches its maximum value, the gas injection stops and therefore foam is no longer generated. The evolution of foam volume as a function of time is shown in Figure 1 for the gases examined.

Foam evolution proceeds in three phases for all gases. (i) Initially, the foam does not drain; the liquid is held in the foam due to capillary suction [32]. The liquid fraction ε is around 20%. During this period the bubbles rearrange to find a stable structure. The effect of the coarsening is relatively small as the critical coarsening time t_c is bigger than the age of the foam (Table 1). (ii) In the second phase, the foam starts to drain due to gravity but the foam volume is constant. The drainage enhances coarsening and the consequent rearrangement of the bubbles [32]. (iii) The third phase is dominated by coarsening and coalescence. The foam volume decreases due to coalescence. The drainage rate declines with time as foam becomes drier. During this stage, black films (common or
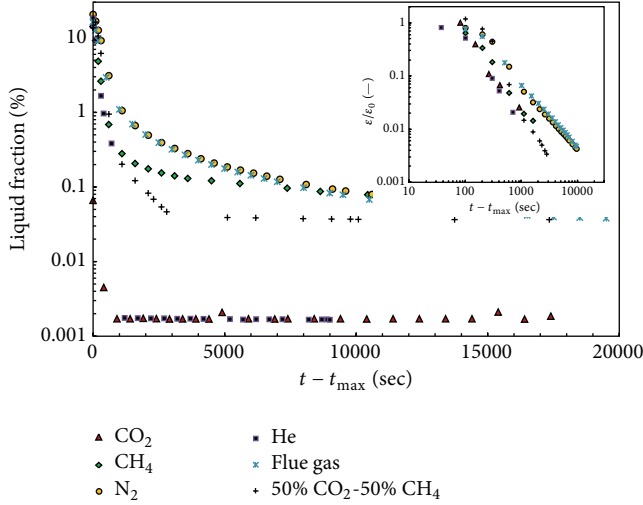
FIGURE 2: Liquid fraction of the foam as a function of time for different gases.

Newtonian) are likely to be formed. The transition from the phases (ii) to (iii) is distinct only for N_2 and flue gas. The transition time between the two stages decreases as the solubility of gas increases. Indeed, with He and CO_2 stages (ii) and (iii) cannot be distinguished. Volume of the foam is inversely related to solubility of the gas in the aqueous solution at any time of the experiment; that is, the lower the gas solubility the larger the volume of the foam. Moreover, addition of insoluble gas to a soluble gas results in more stable foam.

3.2. Effect of Gas Solubility on Drainage. Figure 2 shows the variations of liquid fraction with time in the foams created with different gases. Foams made with helium and carbon dioxide become very dry within few minutes at the stop of foam generation. This leads to very unstable foam. The helium foam height stabilizes within few minutes, not long after the foam becomes very dry at 1000 sec. The behavior of He can be due to the high diffusion coefficient of He, which overcompensates its low solubility in water. Furthermore, we observe from Figure 2 that at the end of the experiment the fraction of liquid in all other gases reaches a similar value, within measurement accuracy of the apparatus.

As an inset of Figure 1, the liquid fraction $\varepsilon/\varepsilon_0$ (normalized by the amount of liquid ε_0 at t_{max}) is presented for all gases on a log-log scale. ε_0 is the initial liquid fraction at $t - t_{max} = 0$. The slopes of $\varepsilon/\varepsilon_0$-time curves are indicators of the rate of liquid drainage from the foam films for each gas and are shown in Figure 3(a). We observe that the drainage rate scales with Henry's solubility constant; that is, the higher the solubility of the gas the higher the rate of drainage. The drainage rate of the foams with mixture of gasses is between the drainage rate of the foams with individual gases.

3.3. Effect of Gas Permeability on Coalescence. The slopes of linear part of foam volume V/V_{max}-time curve (Figure 1) are indicators of the foam decay rate and are shown in

Figure 3(b). As explained in the introduction, the coalescence is driven by the coarsening [27, 29], which depends on the permeability coefficient of the single foam films. The foam decay rate scales with the gas permeability K of the single foam films. Remarkably, the gas mixture (flue gas and CH_4/CO_2) behaves similar to the less permeable gas, respectively, N_2 and CH_4. The only exception in the trend is He. Most of the gases have a critical coarsening time t_c in the range of the foaming time except for He for which t_c is 10 times bigger (see Table 1). CO_2 foams are expected to coarsen during the foaming as t_c is 5 seconds, which is shorter than the foaming time.

The liquid fraction and foam stability seem to be, respectively, influenced by the permeability and solubility of the gas. Thus, the foaming should be a combination of permeability and solubility. This model will be studied in the next section.

3.4. Effect of Gas Solubility on the Foaming. As the foam is generated, gas is solubilized (quantified by Henry's solubility constant k_H). Coarsening can also occur if t_c is shorter than the foaming time. Figure 4 plots the foaming index (FI) of different gasses as a function of Henry's solubility constant. The FI decreases with increasing gas solubility, implying that more gas is required to produce a certain volume of foam as the gas solubility in the aqueous solution increases. Because the surfactant solutions were not saturated with the gas injected prior to the test, the gas dissolves in the solution during the foaming, which decreases the foaming efficiency and the foaming index, accordingly.

To quantify the effect of gas dissolution, the FI is modeled by considering the volume of gas dissolved. Assuming that during the foaming stage the gas fully saturates the surfactant solution (i.e., the gas is dissolved up to the limit given by Henry's law in 50 mL of water), the foaming index can be modeled with a simple equation. Assuming a constant r_f during foaming in (2), the FI_{model} can be approximated by

$$FI_{model} = \frac{V_f}{V_g + V_{dis}}, \qquad (3)$$

where V_g is the volume of gas in the foam, V_f is the final volume of foam, and V_{dis} is the volume of gas dissolved in the 50 mL water calculated with Henry's constant. Defining $FI_0 = V_f/V_g$, corresponding to the FI without gas dissolution (3) becomes

$$FI_{model} = \frac{1}{1/FI_0 + V_{dis}/V_f}. \qquad (4)$$

This equation takes into account the direct effect of gas solubility on the foaming index. Comparing the results obtained from (4) with the foaming index calculated from experimental data (Figure 4) reveals that as the gas solubility increases the mismatch between the model and the data becomes more significant. This indicates that for gas with a high dissolution in the aqueous phase the coalescence rate becomes considerable even in the generation stage.

In order to represent the coarsening, we introduce a volume of the coalesced foam during the generation stage,
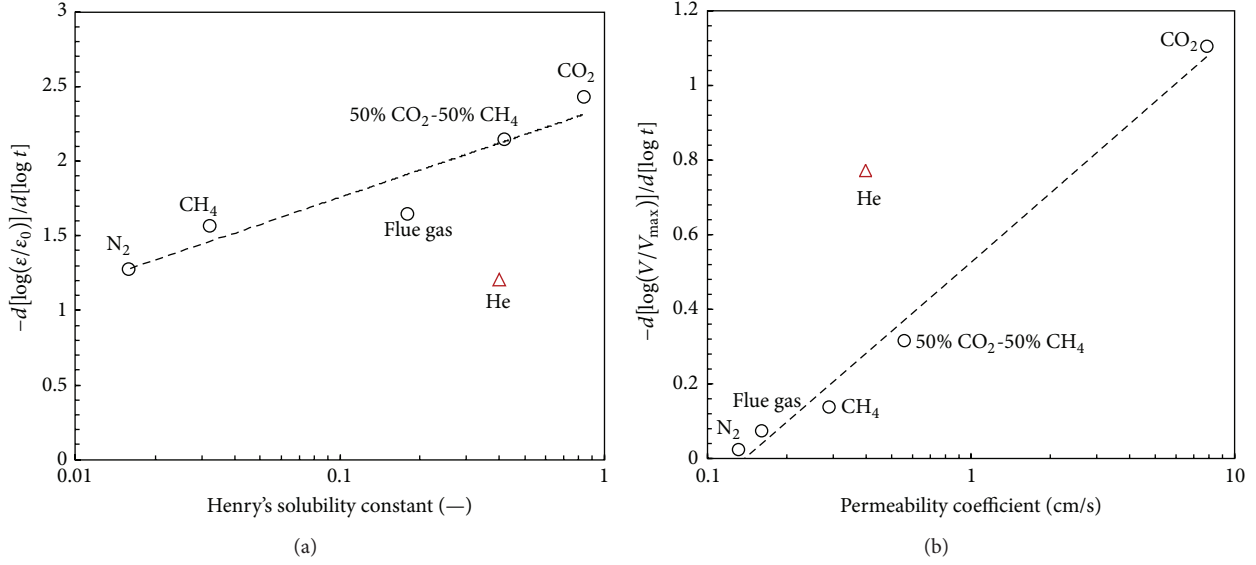
FIGURE 3: Foam drainage versus solubility of the gas and foam decay rate versus gas permeability of single foam films.
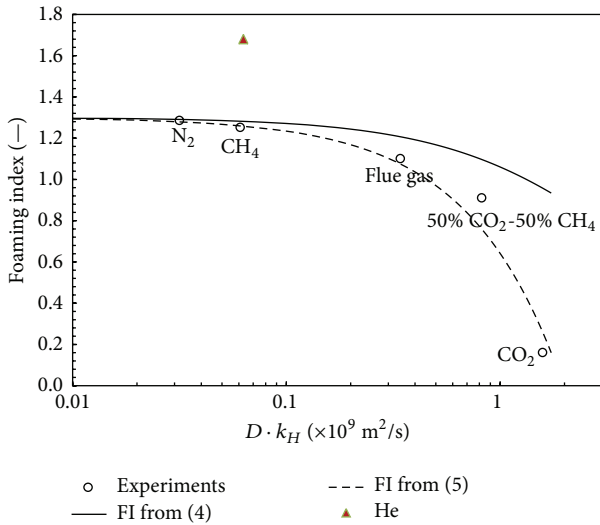

FIGURE 4: Foaming index as a function of gas solubility in the aqueous phase.

V_{coal} represented by the ratio V_{coal}/V_f. We assume that V_{coal} is proportional to the film permeability; that is, $V_{coal}/V_f = \alpha K$. Consequently (4) can be modified to include the foam coalescence in the foaming index:

$$\text{FI}_{model} = \frac{V_f - V_{coal}}{V_g + V_{dis}} = \frac{1 - \alpha K}{1/\text{FI}_0 + V_{dis}/V_f}. \tag{5}$$

The dashed line in Figure 4 is calculated using (5). The fit is obtained by changing the value of constant α to 0.5.

4. Conclusions

A study was conducted to understand the effect of the composition of the gaseous phase on the macroscopic properties of foam. The properties of the foam can be characterized using Henry's solubility constant k_H and the gas permeability coefficient of single foam films, K. The drainage rate increases with increasing gas solubility, while foam-decay rate increases considerably with increasing K. While both drainage and decay rates are influenced when a low-solubility gas is added to a high-solubility gas, the contribution to foam stability is mainly due to reduction in the coarsening rate. With increasing gas solubility the foam coarsening coalescence significantly increases even in the generation stage of foam. This results in poor foaming behavior as the gas solubility in the aqueous phase increases. The foaming index should consider the gas solubility and permeability. Results of helium foam suggest some additional factors must be taken into account in the model.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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