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Microfluidic comparative study of foam flow between a classical and a pH sensitive surfactant

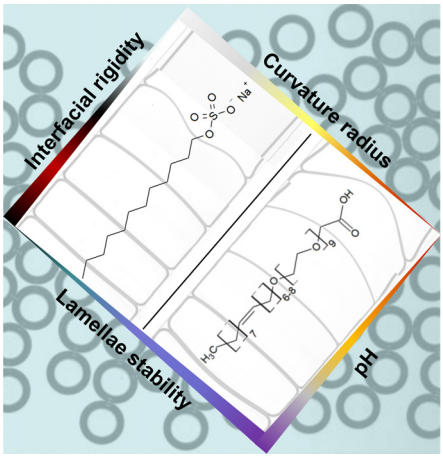
Cyril Micheau, Elisabeth Rosenberg, Loïc Barré, Nicolas Pannacci*

IFP Energies nouvelles, 1-4 Avenue de Bois-Préau, 92852 Rueil-Malmaison Cedex, France

HIGHLIGHTS

- Flowing foam observation and characterization in microfluidic device.
- Comparison between a polyethoxycarboxylate surfactant and sodium dodecyl sulphate surfactant foam stability.
- Foam morphology mainly depends on pressure injection parameters.
- Physico-chemical parameters such as pH highly impact lamellae stability.

GRAPHICAL ABSTRACT



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ABSTRACT

Comparative study between foams made of a pH sensitive surfactant, nonaoxyethylene oleylether carboxylic acid (AKYPO® RO 90 VG) and a classical foaming surfactant, sodium dodecyl sulphate (SDS) was realised using simple microfluidic devices in the scope of Enhanced Oil Recovery (EOR). The influence of injecting fluid parameters such as gas and foaming liquid pressure was investigated and discussed in terms of foam morphology, and lamellae stability and curvature radii. The increasing stability of AKYPO® foam film lamellae by increasing pH is classically explained by the electrostatic repulsive interaction of the double layer in foam film. In other hand, the addition of dodecanol (DOH) on SDS solution seems to have no effect on foam regime and lamellae stability except for high DOH concentration where the foam film shape becomes double bump. The behaviour difference between AKYPO® and SDS lays on the lamellae shape and stability domain. While SDS lamellae stay unchanged with increasing pressure ratio between the gas and the liquid, AKYPO® lamellae curvature radius increases which could be related to surfactants solubilities and structure. These results pointed out the relevance of solution physicochemical parameters on foam stability and flowing and help the understanding of flowing foam in EOR context.

1. Introduction

Liquid foams are non-equilibrium materials [1] composed of a discontinuous gas phase and a liquid phase. Gas is present in the foam in the form of bubbles, spherical in the case of wet foams and

* Corresponding author.
E-mail address: nicolas.pannacci@ifpen.fr (N. Pannacci).

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polyhedral in the case of dry foams. Liquid foams are used in many processes such as ion separation via flotation [2], nuclear decontamination [3] or explosion confinement [4]. A recent application [5] proposes using foam in an Enhanced Oil Recovery (EOR) process in order to extract oil in natural reservoirs. This specific application requires the generation of foams in porous media with the aim of reaching and displacing the oil in place. In porous media, foam's definition is more ambiguous because it is presented as a succession of parallel films, called lamellae, joined together by wetting films on the pore walls [6]. Furthermore, when foams are sufficiently confined, 2D-foams are obtained and their bubbles can distort themselves to adopt a spheroid or cylindrical shape [1,6].

Foam's lamellae are stabilized by surfactant molecules which adsorb at the liquid/gas interface that could change the local surface properties such as surface viscosity, surface tension or elastic moduli [1,7]. These properties directly influence the capacity of the foam to transport gas into porous media which is mainly linked to the foam stability during its generation and flowing.

The higher effective viscosity of foam relatively to gas [8,9] allows to increase the sweep efficiency in porous media and to reduce viscous fingering and gravity segregation due to the low viscosity and the low density of the gas that makes foam a good gas mobility reducing agent [8,10,11]. Moreover, the capacity of foam to accumulate in high-permeability regions reduces gas bypassing by increasing flowing resistance and diverting gas in low permeability pores that improves the EOR process [12,13]. The fundamental understanding of such mechanism is of high importance to better adjust surfactant formulation and extraction process.

Previous works have investigated the physical properties of foam in porous media using lab experiments in sand [14], rocks [10] or micro-device [7,11,15]. Some were interested in the influence of fluid injected parameters on the foam morphology [11] or performance in terms of mobility reduction factor [14], and others on the flowing foam behaviour depending on the porous media geometry [7,11,15]. Less study has been focussed on the influence of physico-chemicals parameters on foam generation, aging and flowing. It is well known that, surfactant composition and dynamics, salt nature and ionic strength or pH [16,17] are of crucial importance on the stability of bulk foams (3D-foams) or films. Moreover, for experiments of foam in porous media, chemical composition of the foaming solution is sometimes modified by salt or dye addition, in order to enhance phase contrast [11,18], or by viscosifying or gelling agent to reduce the foam dynamics (*cf.* aging).

Microfluidic devices are good supports to investigate the influence of physicochemical parameters on foam generation and flowing at the micro-scale. They allowed direct observation of foam with a microscope without using any chemical additive, and a high control level of reproducibility. Coupling with image analysis, and basic image processing, thresholding, and skeletisation of the foam image gives qualitative and quantitative informations on foam generation and flowing.

This work proposes a comparative study between a classical surfactant used as a model surfactant in EOR context, sodium dodecyl sulphate (SDS), [7,8,11,15,18,19] and a pH-sensitive surfactant used in metal separation processes, nonaoxyethylene oleylether carboxylic acid (AKYPO® RO 90 VG) [16,20,21], using microfluidics. Foam morphology and stability are analysed in regard to the injected fluid pressures (N_2 and surfactant solution) and physicochemical parameters such as pH or dodecanol/SDS concentration ratio. Foam phase diagrams are thus obtained and quantified in terms of bubble size, liquid fraction and topology. Foam flow and stability differences between the two selected surfactants are evidenced regarding lamellae stability domain and curvature radius and discussed putting forward interface properties such as surfactant adsorption and surface viscosity.

2. Materials and method

2.1. Microfluidic device

Standard soft lithography in PDMS (PolyDiMethylSiloxane) was used for the microfabrication, whose principle was previously described by Duffy et al. [22] Quasi 2D device with a constant depth (40 μm) was thereby obtained. Bonding of PDMS on glass with plasma cleaner [23] gives hydrophilic channel surface. Due to the reversibility of this treatment *i.e.* channel internal surface becomes hydrophobic after few hours, microfluidic device was renewed for each experiment.

Microfluidic devices used for foam flow experiments are composed of two entrances for gas and surfactant aqueous phase, a cross junction for bubble formation, a chamber for the foam observation of 1200 μm width W and 3000 μm length, and a straight channel (60 μm width) connected to the outside (atmospheric pressure) (see Fig. 1). The depth H of the microchannel is constant and equals to 40 μm.

2.2. Experimental protocol

Entrance pressure above atmospheric pressure was applied for surfactant solution and gas using MCFS™ EZ 7000 mbar (Fluigent). PEEK (polyetheretherketone) flexible tubes of 59.2 cm length and 125 μm inner diameter were used to connect source pressure to gas and liquid inlets. Pressures considered below, named respectively P_{gas} for gas and P_{water} for surfactant solution are upstream applied pressures, above atmospheric pressures. All measurements were carried out when steady flow rate was reached (typically after 30 s). In each experiment, gas pressure drop P_{gas} was kept constant during the flowing (150 mbar or 300 mbar) and the surfactant solution pressure drop P_{water} varies from $4P_{gas}$ to $P_{gas}/60$. Each experiment was performed at least twice. Under such applied pressure and with such aspect ratio of the microfluidic device (1200 μm width W and 40 μm depth H), no deformation of the PDMS cell is observed (no defocusing and no loss of sharpness on microchannel images). This is confirmed by the calculation of the deformation criterion $c = WP/(EH)$, defined by *Gervais* et al. [24], where P is the pressure difference applied on the upper PDMS wall in the observation chamber (the lower glass wall is supposed not to distort) and E is the PDMS Young's modulus ($E \approx 10^6$ Pa). In the case of the maximum water pressure difference applied $P_{water} = 1200$ mbar and considering the hydraulic resistance of the microdevice, the pressure difference applied on the PDMS wall P is found to be equal to 3 mbar, leading to a deformation criterion of 8.97×10^{-3} smaller than one (see calculation in Supplementary material). Accordingly, no deformation is expected at such aspect ratio and such applied pressure.

2.3. Chemicals

Foams were generated by co-injection of nitrogen gas and surfactant solution at around 17.33 mM through the cross junction. The surfactants employed are AKYPO® RO 90 VG (nonaoxyethylene oleylether carboxylic acid, R-O-$(CH_2$-CH_2-$O)_n$-COOH, R = C_{16}/C_{18}, n = 9) from Kao Chemicals (surfactant content 89.0%, M_W = 722 g mol^{-1}) or SDS (Sodium Dodecyl Sulphate, Fischer Scientific, 98%, M_W = 288.38 g mol^{-1}) for the comparative study.

In the case of AKYPO® surfactant solution pH (initial pH at 2.90 at 17.33 mM) was adjusted using either diluted sodium hydroxide solution (NaOH, Titrinorm, 0.1001 N) or nitric acid (HNO_3, Titrinorm, 1.002 N) to reach a pH range between 2 and 12. Surface tension of the different AKYPO® solution in function of pH was obtained from the literature [16] and summarized in Table 1.