# International Symposium on

# HYDROACOUSTICS AND ULTRASONICS

EAA Symposium (formerly 13<sup>th</sup> FASE Symposium) Gdańsk-Jurata, 12-16 May 1997

# Acoustic emission probes of foam drainage processes in H<sub>2</sub>O-C<sub>2</sub>H<sub>5</sub>OH-air systems with content of detergent triton X-100

Rzeszotarska J., Rejmund F., Ranachowski P. Institute of Fundamental Technological Research Problems, PAS Świętokrzyska 21, 00-049 Warsaw

## **ABSTRACT**

The acoustic emission (AE) of short-lived static foams from non-ionic detergent Triton X-100 in ethanol-water solutions has been investigated. The AE of foam drainage process has shown two different stages of behaviour. The first step up to 80 s was without the appearance of AE signals. During this stage the rearrangement of bubbles has been observed. The next, main stage of drainage process proceeded with a high intensity AE generation. The results suggest that the AE technique gives the direct possibility of foam stability measurement.

### INTRODUCTION

Foams of aqueous solutions are the macroscopic homogeneous systems composed of gaseous and liquid phases. One of the more important problems in such polydispersives systems is searching for the method of measuring the resistance of a thin film of liquid to rupture which causes the coalescence process. In foams of low stability hydrostatic pressure and capillary forces cause the drainage of liquid from thin layers to solution under foam. When the drainage process progresses in time the thickness of thin liquid films surrounding the gas inside bubbles decreases and at the critical size a rapid rupture occurs. Meanwhile from the upper layers of bubbles gas diffusion passes across a liquid film to the atmosphere. As a result of these processes the amount of liquid and gaseous phases in foam decreases. At present the growth of bubbles due to diffusion (coarsening) is believed to be the main process determining the lifetime of foam. Diffusion of gas proceeds from smaller to larger bubbles with an average rate of radius growth proportional to square root of time (t 1/2) [1, 2]. Velocity of gas diffusion between bubbles depends on the gas solubility in liquid and on the coefficient of gas diffusion in solution. However, so far there are no mathematical formulas governing the mechanisms of this process.

The problem shows an analogy to the process of grain growth in the polycrystalline materials.

The methods of stability measurement of a static foam in general terms consist in determining the rate value of foam vanishing or in measuring the changes of bubbles size and number in time. For this purpose the following methods are most frequently used: photographic [1], optical [2] and methods based on measuring the pressure over foam [4]. The photographic method consists in the analysis of the sights of frozen foam at different steps of the vanishing process. Optical methods use the change of light transmitted across foam [3] or scattered [2]. The pressure growth in the atmosphere over a foam medium makes it possible to determine the change in area of a thin liquid film in time and thus to determine the velocity of foam vanishing [1].

The methods listed above are not reproducible to a satisfactory degree. This is due to different ways of foam generation. It is believed that the method of foam generation influences the kinetics of an early step of its vanishing. Hence it is necessary to formulate a standard of foam generation.

In this work it was attempted for the first time to use an acoustic emission method (AE) to determine a lifetime of static foam in H<sub>2</sub>O-C<sub>2</sub>H<sub>5</sub>OH-Triton X-100 systems. This AE technique is a direct, non-invasive method of investigating the dynamics of

the bubbles in foam, similar to the method of light scattering [2]. The authors have already used the AE method previously to study the dehydration processes of inorganic salts [5] and the polymorphic transitions of NH<sub>4</sub>NO<sub>3</sub> crystals [6].

## EXPERIMENTAL PROCEDURES

#### A - Materials

The stock solution of Triton X-100 in water with concentration 0.284 mol·l<sup>-1</sup> was prepared a few days before by adding to bidistilled water an appropriate amount of detergent made by Merck. The solutions 0.142 mol·l<sup>-1</sup> of Triton X-100 in the mixture containing 0, 10, 20 and 40 % of ethanol (EtOH) per weight were prepared by dissolving the basic solution with an appropriate amount of ethanol and water. All substances were of analytical grade.

# B - Measurement of viscosity, density, surface tension and propagation velocity of ultrasonic waves

The viscosity of solutions was measured by a capillary viscosimeter with a microprocessor unit made by ECOLAB (Poland). The comparison method was used to measure the times of flow through the capillary  $(\tau_{\scriptscriptstyle 0},~\tau)$  of exactly the same volumes of standard liquid and the investigated one. To measure the density an MG-2 apparatus made by ECOLAB was used. The viscosity in [cP] units was calculated from the formula

$$\eta = \eta_0 \left( \frac{\rho}{\rho_0} \right) \left( \frac{\tau}{\tau_0} \right)$$

The surface tension of solutions was measured by the static method of ring removal from liquid surface. The force at which ring removal takes place is proportional to surface tension value. The values of surface tension ( $\sigma$ ) were determined by the comparison method with respect to the solution not containing Triton X-100.

The velocities of ultrasonic waves were measured with an MPFU apparatus made by ECOLAB. The time at which an ultrasonic wave passes across a sample at a distance of 5 cm was measured. The volume of a cylindrical glass container with the transducers to transmit and receive was 5 ml. The samples were kept at constant temperature with a Peltier thermostat type MRTP.

# C - AE method

The root mean square value of AE signals (RMS) and the sum of RMS were measured with the apparatus shown schematically in Fig. 1. A broadband piezoelectric transducer from the Physical Acoustics Corporation type NANO-30 (Fig. 2) coupled with the bottom of the glass container from outside was used to record the AE signals.

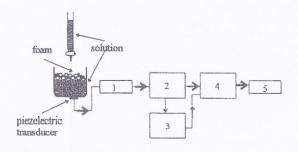


Fig. 1. Diagram of the measuring set-up: 1-preamplifier, 2-acoustic emission analyser DEMA, 3-digital oscilloscope with memory IWATSU OS-6612C, 4-computer, 5-printer.

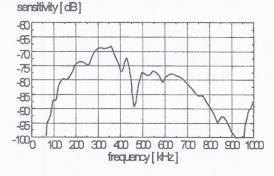


Fig. 2. Frequency response of the broadband piezoelectric transducer type NANO-30 from PAC.

A foam was produced by pouring from a burette 10 ml solution of Triton X-100 into a cylindrical glass container ( $\Phi$ =55 mm) from a height of 400 mm. The diameter of burette outlet was 3 mm. The recording of AE descriptors started 10 seconds after pouring the solution. The AE signals were recorded by an Iwatsu double-beam digital oscilloscope at the following time intervals of foam vanishing: 0-100 s, 100-200 s, 200-300 s, 500-600 s. The AE signals were recorded in the form of 1024 samples with 1  $\mu$ s time interval between two successive samples of the signal, thus the Fourier analysis made it possible to obtain the frequency spectrum of the signal in the range 0-500 kHz. All measurements were made at room temperature.

## RESULTS AND DISCUSSION

Triton X-100 is a well-known, non-ionic surfactant with the chemical formula

$$C_{14}H_{21}O(C_2H_4O)_{10}H$$

It is used as a standard to compare the methods of investigating the dynamics of the surface tension of solutions and the mechanisms of adsorption by the diffusion process at the phase boundary of solution-atmosphere. The critical micelle concentration

Table 1. Physical properties of 0.142 mol·I<sup>-1</sup> Triton X-100 in EtOH-H<sub>2</sub>O solutions

| % weight<br>(EtOH-H <sub>2</sub> O) | % vol.<br>(EtOH-H <sub>2</sub> O) | o<br>[N•m <sup>-1</sup> ]•10 <sup>-3</sup><br>room temp. | σ <sub>s</sub> *) [N•m <sup>-1</sup> ]•10 <sup>-3</sup> room temp. | viscosity<br>η [cP]<br>20 °C | density<br>ρ [g•cm <sup>-3</sup> ]<br>20 °C | velocity of<br>ultrason.wave<br>[m/s]<br>20 °C |
|-------------------------------------|-----------------------------------|--|--|------------------------------|---|--|
| 1                                   | 2                                 | 3  | 4  | 5                            | 6   | 7  |
| . 0                                 |                                   | -  | -  | 1.631                        | 1.005                                       | 1499   |
| 10                                  | 12.36                             | 29.79  | 16.24  | 2.025                        | 0.991                                       | 1566   |
| 20                                  | 24.09                             | 28.68  | 8.85   | 2.795                        | 0.979                                       | 1613   |
| 30                                  | 35.23                             | 28.00  | 4.50   | 3.078                        | 0.961                                       | 1598   |
| 40                                  | 45.83                             | 27.53  | 2.06   | 3.122                        | 0.949                                       | 1537   |

 $^{*)}$   $\sigma = \sigma_{o} - \sigma_{S}$ 

σ<sub>2</sub>-surface tension of solvent (EtOH-H<sub>2</sub>O)

(CMC) of Triton X-100 depends on the method of determination and varies from 1.3 to  $2.7 \times 10^{-4} \text{ mol} \cdot l^{-1}$  [7]. The average molecular weight of micelles which have ellipsoidal shapes is ninety thousands and the average number of aggregates is 143. The intrinsic viscosity of micelles at room temperature ( $\eta$ ) is 5.5 cm<sup>3</sup>·g<sup>-1</sup> [8].

In this work to create foams the solution of Triton X-100 with the concentration 0.142 mol·1¹ i.e. three orders higher than CMC was used. Triton at this concentration exists in the form of micelles. Basic physical properties the water-ethanol solutions of Triton X-100 which influence the rate of thinning liquid film in foam are presented in Table 1. It can be seen from Table 1 that the content of ethanol has a notable effect on the possibility of lowering a surface tension value of the solution by Triton (column 4 in Table 1). The 40 % ethanol concentration lowers the surface tension by about 2 x 10⁻³ N·m⁻¹ while the volume viscosity of the solution increased by about 2 cP as compared with the viscosity of ethanol-water mixtures.

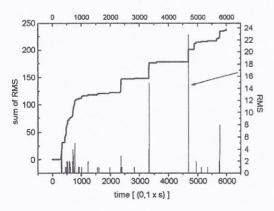
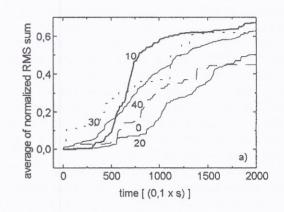


Fig. 3. RMS value and the sum of RMS values recorded during the drainage process of foam produced from Triton X-100 in 10 % EtOH solution.

The counts rate, events rate, maximum amplitude of the acoustic emission signals and the root square value (RMS) were recorded using an acoustic emission analyser of DEMA type produced at the Institute of Fundamental Technological Research. Among the descriptors of AE signals known in the literature [9], the recorded RMS values were carefully examined in this work.



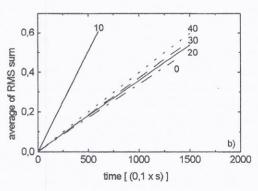


Fig. 4. Dependencies: a) normalized sum of RMS value as a function of time for solutions of Triton X-1000 with 0, 10, 20, 30, and 40 % of EtOH; b) linear approximation of two stage processes from Fig. 4a).

Table 2. Drainage dynamics of foam in X-100-EtOH-H<sub>2</sub>O system.

| %<br>EtOH | average time<br>to collapse<br>[s] | % of AE energy<br>emitted during<br>stage 2 of the<br>process | duration of<br>stage 2<br>[s] | velocity of AE<br>energy emission<br>at stage 2<br>[%/s] |
|-----------|------------------------------------|---|-------------------------------|--|
| 0         | 80                                 | 44  | 140                           | 0.34   |
| 10        | 40                                 | 60  | 99                            | 1.00   |
| 20        | 50                                 | 44  | 160                           | 0.36   |
| 30        | 40                                 | 60  | 130                           | 0.37   |
| 40        | 10                                 | 58  | 160                           | 0.40   |

The RMS level of AE signals is proportional to the acoustic energy radiated from the sources of the acoustic waves during the process of foam destruction. An example of the RMS value and the RMS sum as a function of time recorded during the collapse process of foam created from the 10 % per weight EtOH solution is presented in Fig. 3.

The dependencies of RMS and RMS sum give a faithful representation of the dynamics process of bubbles destruction in Triton X-100-EtOH-H<sub>2</sub>O systems. The investigated processes of foam vanishing while studying the dynamics of RMS descriptor can be divided into three stages. Stage one extends to 80 seconds (Table 2.) and does not show the appearance of AE signals. The structural changes of foam are performed without the rupture of bubbles. Duration of the first stage of the process is related to physico-chemical properties, especially the viscosity of solution and the solubility of air in the EtOH-H<sub>2</sub>O mixtures. The length of time to collapse initiation of foams decreases together with a decrease of ethanol concentration in thin liquid films.

During stage two extending from 100 to 160 seconds the destruction of foam runs "explosively". About 50 % of acoustic energy is emitted in the form of signals with a frequency in the range 20-250 kHz. The velocities of AE energy emission at this stage (Table 2) were determined from the dependencies presented in Fig. 4b. The rates of foam destruction for the solutions 0, 20, 30 and 40 % are adjacent. The high rate of foam destruction for the solution of 10 % concentration is difficult to explain. During stage three of the destruction process the acoustic emission became weaker, and rupture of bubbles occurred rarely. During the period of the last 100 s on the average four AE signals were recorded. The signals were emitted by the largest bubbles of the longest time of existence.

### CONCLUSIONS

(1) The acoustic emission measurements of shortlived static foams can serve to determine the properties of decay process.

- (2) Both the RMS value and the average frequency spectra have shown the strongest acoustic activity during an interval of the first 100 s. AE activity increases with increasing ethanol concentration in solution.
- (3) The analysis of average spectra at the selected intervals of foam decay process have revealed that the low frequency peaks are generated by the rupture of bubbles while the high frequency peaks are due to other sources of foam decay.

This work was partly supported by the KBN Grant Nr 7T07B 02008.

#### REFERENCES

- Prud'homme P.K., Khan S.A., (1996), Foams: Theory Measurements and Applications, Surfactant Science Series Vol. 57, Marcel Dekker Inc., New York
- Durian D.J., Weitz D.A., Pine D.J., (1991), Multiple Light-Scattering Probes of Foam Structure and Dynamics, Science, 252, 686-688
- 3. Ross S., Cutillas M.J., (1955), The Transmission of Light by Stable Foams, J. Phys. Chem., 59, 863-866
- Princen H.M., (1988), Pressure /Volume/ Surface Area Relationships in Foams and Highly Concentrated Emulsions: Role of Volume Fraction, Langmuir, 4, 164-169
- Rzeszotarska J., Rejmund F., Ranachowski P., (1995), Differential Thermal Analysis and Acoustic Emission of Powdered Salts of CuCl<sub>2</sub>·2H<sub>2</sub>O and Cu(ClO<sub>4</sub>)<sub>2</sub> Evoked by Temperature Changes, Ceramics, 47, 101-107
- Rzeszotarska J., Rejmund F., Ranachowski P., (1997), Acoustic Emission of Polymorphism in Ionic Crystals of Ammonium Nitrate (I-IV), Acoust. Letters, in print
- Perkowski J., Mayer J., Ledakowicz S., (1995), Determination of Critical Micelle Concentration of Non-ionic Surfactants Using Kinetic Approach, Colloids Surfaces, 101, 103-106
- Robson R.J., Dennis E.A., (1977), The Size, Shape, and Hydration of Nonionic Surfactant Micelles. Triton x-100, J. Phys. Chem., 81, 1075-1078
- Soulsbury K.A., Wade A.P., Sibbald D.B., (1992), Rules-Based Approach to Classification of Chemical Acoustic Emission Signals, Chemometrics and Intelligent Laboratory Systems, 15, 87-105