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# Microrheometric study of damage and rupture of capsules in simple shear flow

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## Summary

Capsules under shear flow undergo fatigue phenomena which may lead to rupture. We considered bioartificial microcapsules made of ovalbumin reticulated with terephthaloyl chloride (TC) with the objective to characterize the impact of damage on the mechanical properties of the membrane. Moreover, we conducted a statistical study of the occurrence of rupture depending on the capsule radius. The break-up probability increases with the capsule radius.

## 1. Introduction

Capsules, which are liquid droplets enclosed in a thin elastic membrane, are present in nature (red blood cells, phospholipid vesicles) and in various industrial applications (biotechnology, pharmacology, cosmetics...). They are used to protect and transport active ingredients by isolating them from the external fluid suspension.

When placed in an external flow at high shear rates, capsules undergo deformation and damage. Rupture occurs when the capsule membrane loses its rigidity at some locations, making it incapable to resist any longer. Depending on the applications, capsule damage is to be prevented to preserve the encapsulated substance or on the contrary provoked to initiate the rupture and then the release of the inner fluid. It is thus necessary to understand the mechanisms responsible for damage and the parameters that control rupture initiation.

Numerous numerical studies have been conducted on the deformation of capsules [1, 2] by implementing a fluid-structure interaction model accounting for the membrane large deformations induced on a capsule subjected to an external flow. Recently, a damage model has been developed by Grandmaison et al [3] that accounts for the initiation and growth of microdiscontinuities and subsequently the local initiation of rupture. On the experimental side, very few studies have been carried out on the rupture of capsules. Chang & Olbricht [4] showed the capsule burst in a simple shear flow at high shear by membrane fatigue. In their study, the break up was initiated from one of the major axis tips of the deformed capsule and propagated in the shear plane. Rehage & Koleva [5] also observed the rupture of thin polysiloxane microcapsule under simple shear flow. The rupture occurred in the central region, close to the tips of the flow vorticity axis (zones of maximum tension) at small deformations. Rupture was also observed in hyperbolic flow by Xie & al [6] using Chitosan/PFacidYN thin membrane. When the hydrodynamic stress exceeds the limit for breakup, the microcapsule stretches up leading to the formation of two lobes connected by a thin filament which is then destabilized in droplets.

None of the experimental studies carried out have investigated capsule damage phenomena. However, this is a crucial point in order to control rupture and build a reference set to validate the numerical damage models. To fill this gap, the objective of the present work is

dedicated to a rheometric study of the capsule damage and rupture based on an identification of the membrane mechanical properties. A decrease in the capsule surface shear modulus will inform on its damage and upcoming rupture. After having detailed the experimental setup and methods in Section 2, we present the results of the parametric identification of capsule damage in Section 3 and a statistical study of the rupture is presented in Section 4.

## 2. Experimental setup and methods

### 2.1. Capsule fabrication

Microcapsules are prepared by means of interfacial cross-linking of an emulsion of 1 ml of an aqueous solution, consisting of 10 % (w/v) ovalbumin (A5253-250G, Sigma) dissolved in phosphate buffer (P3619-1GA, pH=7.4, Sigma), mixed with 10 ml of vegetable oil (ISIO 4, Lesieur) using a laboratory vortex (Heidolph TopMix 94323). Then, 10 ml of vegetable oil containing 2.5 % (w/v) terephthaloyl chloride (120871-250G, Sigma) is added to the emulsion. The interfacial reticulation time is 15 min. Once prepared, capsules are rinsed before use and suspended in glycerol at a concentration of 2 %. Owing to concentration differences leading to osmotic effects, the capsules are pre-stressed with an inflation ratio  $\alpha = \frac{a}{a_0} - 1 = 53\%$ , where  $a_0$  is the reference capsule radius and  $a$  the pre-stressed capsule radius.

### 2.2. Microrheometry

Our study of capsule motion is carried out in a Couette flow using two counter-rotating concentric cylinders as illustrated in figure 1 and between which we introduce a volume of 12 ml of capsules suspended in glycerol. The distance between the two cylinders is about 1 mm. After the capsules are placed in the region between the cylinders and the flow is started for a chosen shear rate, their motion and deformation are recorded by a video camera (model LM165M, Lumenera) mounted below the outer cylinder to provide a bottom view of the capsules. The capsule image is inverted by passing through a 45° mirror (figure 1). To enhance the resolution of the recorded images, a spot light (KL 1600 LED, SCHOTT) is used.

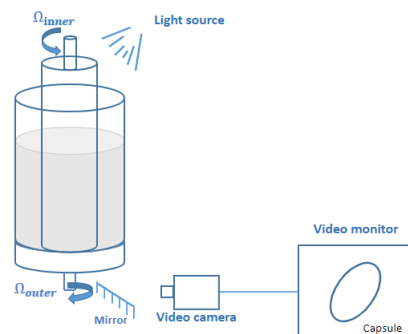


FIGURE 1 – Schematic of the Couette apparatus.

### 2.3. Identification of the membrane elasticity

To identify the surface shear modulus  $G_s$  of the capsule membrane, an experimental-numerical strategy is used. From the experience, we know the values of the shear rate  $\dot{\gamma}$ , the viscosity  $\mu$  of the capsule-in-glycerol suspension and the contour of a deformed capsule observed during the shear flow which is obtained manually using ImageJ. A least squares fit of the contour with an ellipse provides the values of the semi-axes  $L_1$  and  $L_2$  and then the Taylor parameter  $D_{12} = \frac{L_1 - L_2}{L_1 + L_2}$ . From previous numerical simulations of capsule deformation [1] we exploit the plot of variation of  $D_{12}$  with the capillary number ( $Ca = \frac{\mu \dot{\gamma} a}{G_s}$ )  $D_{12} = f(Ca)$  and the plot  $L_3/a = g(D_{12})$ , where  $L_3$  is the third semi-axis of the deformed capsule [7]. From

$D_{12} = f(Ca)$  we obtain  $Ca$ .  $L_3/a$  is obtained from  $L_3/a = g(D_{12})$ . The capsule radius  $a$  is obtained applying the volume conservation of the capsule :  $a = \sqrt[3]{L_1 L_2 L_3}$ . Finally the surface shear modulus  $G_s$  is obtained through the expression  $G_s = \frac{\mu \dot{\gamma} a}{Ca}$ .

## 2.4. Probability of rupture

The capsule distribution in glycerol is considered to be homogeneous in this study. To identify the number of ruptured capsules, we quantify their number in a volume of  $10 \mu\text{l}$  of the capsule-in-glycerol suspension, we shear the mixture using the microrheometer and we quantify the number of sheared capsules. We assume that the disappeared capsules are ruptured during the flow. The results of this study are presented in section 4.

## 3. Capsule damage

This section aims at presenting the results obtained in terms of the evaluation of the surface shear modulus  $G_s$ . We study its dependence on the size of the capsules and its evolution as a function of the exposure time to shear. To this end, we evaluate  $G_s$  and the capsule radius following the method presented in 2.3 for different exposure times  $t_E$ .

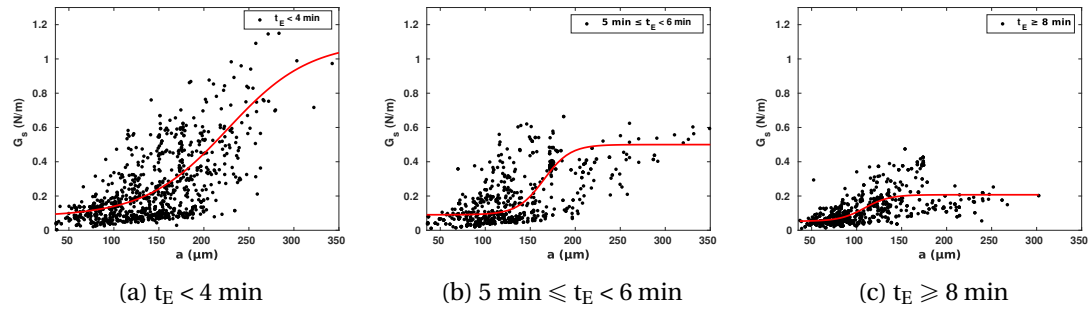


FIGURE 2 –  $G_s$  evolution as a function of the capsules radius for different exposure times  $t_E$

We present in Figure 2 the evolution of  $G_s$  as a function of the capsules radius for different exposure times. We show that  $G_s$  increases with the capsule radius and adjusts to a sigmoid function. When the exposure time to shear  $t_E$  exceeds 4 minutes, we observe a progressive decrease of the surface shear modulus for capsules of radius larger than  $60 \mu\text{m}$ . In figure 3, we present specifically the evolution of  $G_s$  as a function of the exposure time for capsules of specific sizes. We attribute this decrease in surface shear modulus to the degradation of the mechanical properties of the membrane.

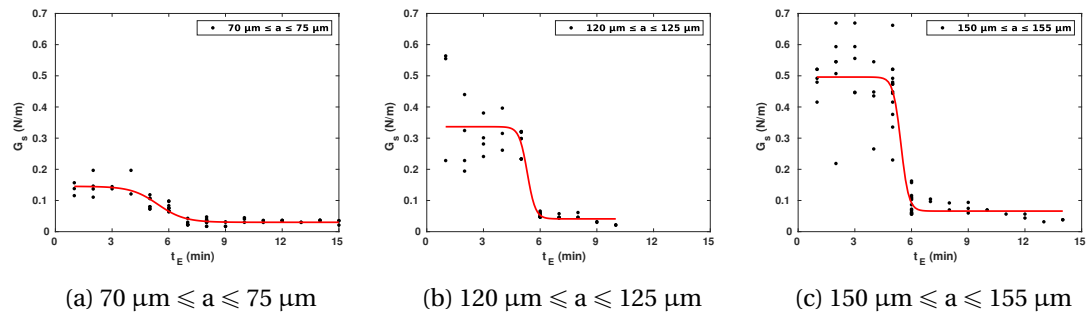


FIGURE 3 –  $G_s$  evolution as a function of the exposure time to shear for three sizes of capsules

## 4. Capsule rupture

To obtain the percentage of ruptured capsules, we quantify their number before and after exposure to shear suspending them in simple shear flow for different exposure times  $t_E$ . In this study, we only present the results obtained for  $t_E = 10$  min. The capsules no longer present at time  $t_E$  are considered broken. In figures 4 and 5, we present the results obtained for capsules sheared at a rate of  $300 \text{ s}^{-1}$  for 10 min and a viscosity of  $0.95 \text{ Pa}\cdot\text{s}$ . As shown in figure 5, the percentage of disappeared capsules increases with their radius until their total disappearance for capsules having radius larger than  $60 \mu\text{m}$ . This observation provides information on rupture kinetics. The capillary number of broken capsules (ratio of the viscous to the elastic forces), evaluated using equation (2) ranges from 0.12 to 0.2.

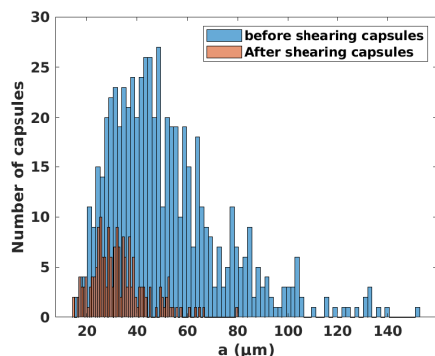


FIGURE 4 – Number of capsules in a volume of  $10 \mu\text{l}$ .

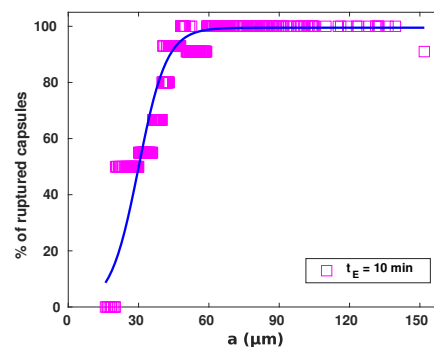


FIGURE 5 – Pourcentage of ruptured capsules as a function of their radius.

## 5. Conclusion

The study of damage is crucial to control and prevent capsule rupture. To be predictive, the damage models used in the simulations [3] must be identified. To this aim, we propose a parametric method based on the membrane mechanical properties evolution. Such an identification is a crucial point for the evaluation of the parameters of the damage models. The second point explored in this work is based on a statistical study of rupture kinetics depending on capsule radius.

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