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

Chaymae El Mertahi, Nicolas Grandmaison, Claire Dupont, Rachid Jellali, Anne-Virginie Salsac*

Université de Technologie de Compiègne, CNRS, Biomechanics and Bioengineering, 60203 Compiègne, France

Delphine Brancherie

Université de Technologie de Compiègne, Roberval, 60203 Compiègne, France

*Corresponding author. Email: anne-virginie.salsac@utc.fr

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1. Introduction

Microcapsules are fluid droplets surrounded by a thin elastic membrane, used in industry to protect and transport active substances, aromas, cells When subjected to an external flow, they undergo strong deformations, which can potentially lead to their breakup. Depending on the application, the rupture is to be avoided to preserve the internal content or promoted to allow the release of the internal substance. To control this release, it is crucial to understand the mechanisms of damage and rupture.

Since the 1990's, capsule membrane properties have been studied until breakup, subjecting the particles to various flow conditions: simple shear flow (Chang and Olbricht 1993), hyperbolic flow (Xie et al. 2017), spinning flow (Husmann et al. 2005), tube flow (Wang et al. 2021).... However, few studies have provided a comprehensive set of experimental data on rupture or has investigated the underlying damage phenomena leading to breakup. To overcome the lack of experimental data in the literature, we have conducted a microrheometric study of microcapsules, subjecting them to simple shear flow conditions leading to their damage and rupture.

2. Methods

2.1 Suspension preparation for microrheometric tests

2% (w/v) ovalbumin capsules are prepared using an interfacial cross-linking technique (Edwards-Lévy et al. 1993). This method yields spherical, deformable

capsules with radii spanning from a few microns to a few hundreds of microns. To prevent osmotic stress due to concentration differences between internal and external fluids, capsules are suspended in a glycerol solution that contains 3% (w/v) ovalbumin.

2.2 Microrheometry

Our study of capsule motion is conducted in a Couette flow, produced within a counter-rotating micro-rheometer (Wang et al. 2021). A volume of 10 ml of capsule-in-glycerol suspension is introduced into the 1 mm-gap between the cylinders and sheared varying two parameters: the shear rate $\dot{\gamma}$ and the exposure time t_E during which capsules are subjected to shear. The profile of deformed capsules is captured from below using a 45° mirror placed under the inter-cylinder gap. The characteristic lengths are then determined by fitting the profiles to an ellipse with ImageJ, giving the semi-major axis L_1 and the semi-minor axis L_2 of the capsule.

2.3 Identification of the membrane elasticity and its evolution

To determine the elastic properties of the capsule membrane, we use the numerical results from Wang et al. (2021), linking the capsule deformation in the shear plane ($D_{12} = \frac{L_1 - L_2}{L_1 + L_2}$) to the elastic capillary number Ca (ratio of the viscous to elastic forces) and to \tilde{E}_3 (the semi-axis length of the capsule in the direction perpendicular to the shear plane, normalized by the initial radius of the capsule a). From the numerical curves obtained with the neo-Hookean law, we determine the values of Ca and \tilde{E}_3 . We can then deduce the values of the initial capsule radius $a = \sqrt{L_1 L_2 \tilde{E}_3}$ from volume conservation and of the surface shear modulus $G_s = \frac{\mu \dot{\gamma} a}{Ca}$ from the capillary number definition, where μ is the average viscosity observed during the experiment, derived from rheometer measurements taken at each time point. The viscosity varies across experiments due to its sensitivity to temperature changes.

2.4 Experimental characterization of capsule rupture

In this study, we assume that the distribution of capsules in glycerol is homogeneous and that the occurrence of rupture directly impacts the number of capsules present in the solution. To characterize the capsule rupture, we thus observe 10 μ l of the capsule-in-glycerol suspension under an optical microscope before exposure to shear and we manually count the number and measure the size of capsules present in the sample. The same procedure is applied on 10- μ l-samples taken after exposure to shear. We vary the values of shear rate and exposure time to shear t_E , and determine the percentage of ruptured capsules by

comparing the capsule number before (N_{before}) and after (N_{after}) shearing.

3. Results and discussion

3.1 Capsule damage

We explore damage by examining the time evolution of capsule surface shear modulus G_s in flow. Our study demonstrates that when t_E exceeds 2 minutes, a notable decrease in surface shear modulus occurs. This underlines the significant role of hydrodynamic forces in causing capsule damage.

3.2 Capsule rupture

In this study, we investigate the influence of various parameters on rupture dynamics, particularly focusing on capsule size and capillary number. We find a clear correlation between capsule size and rupture kinetics, noting that larger capsules tend to rupture more rapidly. Additionally, our analysis shows that as the capillary number increases, the percentage of ruptured capsules ($\Delta N = \frac{N_{before} - N_{after}}{N_{before}}$) also rises. This observation can be attributed to the dominance of viscous forces over the capsule's elastic resistance at higher capillary numbers, increasing the probability of rupture.

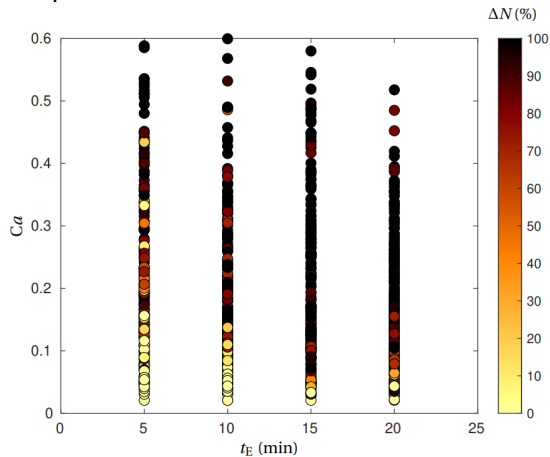


Figure 1. Evolution of the percentage of ruptured capsules as a function of the capillary number and exposure time to shear.

Furthermore, we examine how exposure time t_E affects rupture behavior. Our analysis shows that as exposure time increases, the capillary number needed for rupture decreases (Figure 1). This phenomenon is attributed to the progressive accumulation of residual stresses within the capsule over time due to prolonged exposure to shear forces. These accumulated stresses weaken the capsule's membrane, reducing the limit stress required for rupture.

4. Conclusions

During our study on capsule damage and rupture under shear flow, we have made several significant observations. Firstly, we have noted a gradual decrease in surface shear modulus over time, indicating decreased mechanical integrity with prolonged shear exposure. This underscores the importance of understanding the evolving material properties under shear stress. Moreover, we have found that rupture resistance is influenced not only by the capillary number but also by factors such as exposure time, shear rate, and capsule size. Higher capillary numbers and shear rates have resulted in faster rupture, while larger capsules have tended to rupture earlier.

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Conflict of Interest Statement

The authors report no conflict of interest.

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