Deformation, orientation and bursting of microcapsules in simple shear flow: Wrinkling processes, tumbling and swinging motions

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A Unverfehrt, I Koleva and H Rehage

1 Technical University Dortmund, Department of Physical Chemistry, Germany
2 Phoenix CBS GmbH, Bad Blankenburg, Germany

anja.unverfehrt@tu-dortmund.de, ivanka.koleva@tu-dortmund.de, heinz.rehage@tu-dortmund.de

Abstract. In a series of experiments we studied the deformation and orientation behaviour of microcapsules in simple shear flow. For a large number of capsules we observed folding processes which were induced by the bending resistance, by membrane pre-stresses or the mechanical asymmetry of the surrounding viscoelastic wall materials. Periodic oscillations of the inclination angle were detected for non-spherical particles. At low shear rates a tumbling motion occurred in which the capsule turned around its axis. A swinging mode at evaluated shear rates was accompanied by tank-treading motions, a rotation of the membrane around the capsule core. Between these two well-known motions we also observed an intermittent regime.

1. Introduction

Microcapsules with well-defined wall materials are widely used in technical, cosmetic and pharmaceutical applications. There is still a great demand for new designs of membranes with controlled release properties and bursting processes. Taylor-made capsules require, therefore, the knowledge of their mechanical properties. In scientific experiments capsules are not only used as transport carriers for active ingredients and substances but also as artificial model systems for biological cells, e.g. Red Blood Cells (RBC). In order to get more information on the flow induced deformation of biological cells, we investigated microcapsules in simple shear flow. For that matter the complex RBC membrane was replaced by a simple structured polymer material with adjustable viscoelastic properties. This procedure allowed to study basic principles and mechanisms with variable parameters.

A microcapsule generally consists of a liquid core enclosed by a deformable membrane. These particles are often synthesized by interfacial polymerization of emulsion droplets. The dynamic response of microcapsules in simple shear flow depends on many parameters. The initially shape [1], the viscosity ratio of the inner and outer phases [2], the membrane constitutive laws [3], the bending stiffness [4-7] and the membrane pre-stresses [8] have striking influence on the deformation and orientation behavior. In former experiments, we observed wrinkling instabilities for polysiloxane and polyamide microcapsules [9-11]. Osmotic pressures, membrane pre-stresses and the bending elasticity might be responsible for these phenomena [12-14].
A model for initially spherical microcapsules in simple shear flow was first proposed by Barthès-Biesel [15]. A capsule with a membrane of negligible bending stiffness is deformed by hydrodynamic forces to an ellipsoid and this particle will be orientated at a constant angle of $\Theta = 45^\circ$ (figure 1).

The deformation is described in terms of the Taylor-Parameter $D$, in which $L$ and $B$ represent the major and minor axis of the elliptically deformed microcapsule.

$$ D = \frac{L - B}{L + B} \quad (1) $$

The surface Young’s modulus $E_s$ of the enclosing, viscoelastic shells can be determined using Hooke’s law for small elastic deformations or Mooney-Rivlin’s law for flexible membranes which exhibit non-linear elasticity. For purely elastic membranes, however, a linearity between the Taylor-parameter $D$ and the shear rate $\dot{\gamma}$ was detected in regime of small deformations. This enables the calculation of $E_s$ using the simple equation (2) [16,17]. Here $\eta_a$ denotes the viscosity of the outer phase, $r$ the capsule radius and $C_a$ the capillary number.

$$ D = \frac{25\eta_a r \dot{\gamma}}{4E_s} = \frac{25}{4} C_a \quad (2) $$

Initially non-spherical capsules show an interesting orientation behavior. It depends mainly on the shear rate, the capsule shape and the viscosity ratio $\lambda$ of the inner and outer fluids. At low shear rates we observed a “tumbling motion” (TU), in which the inclination angle varied continuously between $+90^\circ$ and $-90^\circ$ [1,2,11]. The “swinging motion” (SW) was detected at evaluated shear rates and characterized by oscillations of $\Theta$ around positive values [1,2,11,18]. This mode was superimposed by a “tank-treading” (TT) motion. Tank treading describes the rotation of the membrane around its core. The tumbling motion and the swinging process were accompanied by periodic shape oscillations [1,2,9-11] The influence of the viscosity ratio was analyzed in model recently proposed by Keller and Skalak [19]. These authors observed that less viscous particles undertook a “tank treading” motion (TT) while more viscous ellipsoids tumbled. Skotheim and Secomb extended this model and included the “shape memory effect” [20]. They predicted that the transition from tumbling to swinging occurs via an intermittent regime (IR), in which a series of oscillation is interrupted by a tumble. Sui et al. presented a numerical simulation which also suggested the presence of an intermittent regime, but there was not sufficient evidence to confirm this special motion [1]. Recent studies of Yazdani et al. took the membrane viscosity of capsules into account [21]. They observed that a transition from SW to TU occurred when the membrane viscosity increased. Additionally they concluded that membrane viscosity had similar effects as the inner viscosity of the capsule.

In contrast to this theoretical work, less experimental results are published up to now [9-11,18,22,23]. To support these theoretical and numerical models we investigated microcapsules in simple shear flow by means of an optical flow cell (rheoscope).
2. Experimental

The capsule membranes were synthesized by interfacial polycondensation using self-assembled monolayers (SAM) of octadecyltrichlorosilane (OTS). In contact with water the oil soluble OTS molecules hydrolyzed and formed the corresponding alcohols (silanols).

$$\text{Si-Cl} + \text{H}_2\text{O} \rightarrow \text{Si-OH} + \text{HCl}$$

The formed alcohol was even more hydrophilic and adsorbed at the oil/water interface. In a second step silanol molecules condensed forming siloxanes. Silanols could also react with the origin octadecyltrichlorosilane molecules.

$$2 \text{Si-OH} \rightarrow \text{Si-O-Si} + \text{H}_2\text{O}$$

$$\text{Si-Cl} + \text{Si-OH} \rightarrow \text{Si-O-Si} + \text{HCl}$$

As OTS had three functional groups, cross-linking processes occurred, and we obtained a stable polysiloxane network with viscoelastic properties. The reaction took place at room temperature and slightly alkaline or acid conditions.

The polysiloxane network was characterized at the water/oil-interface by means of two-dimensional rheology. In these experiments we obtained basic information on the kinetics of surface gelation, the network type and the viscoelastic regime (LVR). The microcapsules were studied in an optical flow cell (rheoscope). For each experiment we used separate conditions. For interfacial shear rheometers the density difference should be high to give a planar interface. In an optical flow cell however, the densities of the organic and the water phase should be nearly the same in order to avoid sedimentation and creaming of capsules. These different conditions were realized by using mixtures of organic and polar solvents. To compare the results of the flat and curved polysiloxane membranes we adjusted the OTS bulk concentration in such a way that the interfacial monomer concentration in the different solvent mixtures was identical. We determined the mean surface area of the flat membranes and the microcapsules with diameters of 300 µm and adjusted the interfacial excess concentration $\Gamma$.

$$\Gamma = \frac{c \cdot V \cdot N_A}{A} \quad (3)$$

$N_A$ denotes the Avogadro constant, $V$ represents the volume of the organic phase and $c$ denotes the bulk concentration of OTS. Equation (3) holds as long as all monomers adsorb at the interface and become part of the two-dimensional network. In previous experiments, we have shown that this condition is approximately fulfilled [23,24].

Relevant data of the volume and surface concentrations are summarized in Table 1.

<table>
<thead>
<tr>
<th>$\Gamma$ / molecules nm$^{-2}$</th>
<th>$c_{\text{OTS (flat networks)}}$ / mM</th>
<th>$c_{\text{OTS (capsules)}}$ / mM</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.0</td>
<td>0.00137</td>
<td>0.2</td>
</tr>
<tr>
<td>12.0</td>
<td>0.00274</td>
<td>0.4</td>
</tr>
</tbody>
</table>

2.1. Flat membranes

The interfacial shear rheometer was based on a conventional rotational ARES-spectrometer (TA instruments). The ultra-thin membranes were investigated using a 2D-Couette-geometry. The outer cylinder consisted of a glass cup attached to the motor unit. A biconical titanium plate served as the measuring tool and was placed exactly at the interface between oil and water. 30 ml of the aqueous phase, consisting of 90 wt% glycerol and 10 wt% 1 M NaOH-solution ($\rho = 1.241$ g/cm$^3$), was poured
into the glass cup. It was covered with 10 ml of an OTS solution dissolved in a mixture of p-xylene and 1,2,4-trichlorobenzene with a density of 1.1 g/cm³.

The two dimensional storage modulus $\mu'$ and the loss modulus $\mu''$ were calculated from experiments with sinusoidal oscillations. Three different measurements were carried out successively: A time-sweep-test to observe the process of surface gelation, a frequency-sweep test to get information of the network-type and a strain-sweep-test to determine the viscoelastic regime.

2.2. Synthesis of microcapsules

In order to synthesize microcapsules with spherical shapes and equal sizes, we used a microfluidic device with T-shaped channel. In this instrument a disperse oil phase was injected vertically to the flow direction of a continuous aqueous phase. Due to the acting shear forces tiny oil droplets were generated which contained the reacting OTS monomers. The aqueous phase consisted of 90 wt% glycerol and 10 wt% of an aqueous 1 M NaOH-solution ($\rho = 1.241$ g/cm³). To adjust the same density for the oil phase, OTS was solved in a mixture of p-xylene and 1,2,4-trichlorobenzene ($\rho = 1.243$ g/cm³). We used automatic syringes in order to generate a constant solvent flow through the PVC-tube. We succeeded in producing capsules with diameters of 300 µm for aqueous solution velocities of $v_a = 85$ ml/h ($d_{\text{needle}} = 2\text{mm}$) and organic solution velocities of $v_i = 0.3$ ml/h ($d_{\text{needle}} = 85\mu\text{m}$). The polymerizing droplets were collected in a beaker where they remained stored until the membrane was formed. The density adjustment and the high viscosity (261 mPas) of the continuous phase prevented the capsules from sedimentation, coalescence and creaming processes. This procedure leaded to nearly monodisperse and spherical particles. A very small deviation from the true spherical shape was caused by the flow processes in the tube. After different polymerization times the microcapsules were inserted into the optical flow cell.

2.3. Optical flow cell (rheoscope)

The optical flow cell (figure 2) consisted of a coaxial cylinder system which was inserted into an inverse microscope. Due to the different radii with 42.4 and 41.0 mm a gap of 1.4 mm was formed. We induced a laminar shear field by rotating the cylinders in opposite directions. In the centre of the annular gap a stagnation zone was formed where the microcapsules did not translate. Each cylinder was connected to a DC motor drive instrument and could be controlled separately. For measurements an automatic control unit adjusted the shear rate and retained the capsule within the field of observation. The outer cylinder had glass windows to observe the particles within the gap. To avoid turbulences in simple shear flow a small Reynolds number was required. We therefore filled the gap with a highly viscous solvent, and all experiments were carried out in anhydrous glycerol.

![Figure 2. Schematic drawing of the rheoscope: 1- inner cylinder, 2 - outer cylinder, 3 - motor drive unit, 4 - belts, 5 - microscope objective, 6 - light source.](image)

The microcapsules produced in the microfluidic device were placed via a syringe into the centre of the gap. In a series of experiments, we performed measurements with increasing, decreasing and constant shear rates. The capsule’s motion during flow was analyzed by means of a high speed camera (Kodak SR-Corder Motion Analyzer). A video file with 25 images/s and a log file with the value of the shear rate were recorded simultaneously. The capsule deformation and the inclination angle were calculated from the capsule contour and position using the NI VDM software. More details, concerning the optical measurements are summarized in [25].
2.4. Materials
As octadecyltrichlorosilane is extremely reactive towards small amounts of water we used anhydrous solvents. P-xylene (99+%) and 1,2,4-trichlorobenzene (99+%) were purchased from Sigma-Aldrich and stored under argon. Their water content was determined by Karl-Fischer-Titration to 0.0025 wt% H₂O. OTS (ABCR) was purified via vacuum distillation (7.5 *10⁻² Torr, 155°C). Anhydrous glycerol was purchased from Merck.

3. Results and discussion

3.1. Interfacial shear rheology of the planar polysiloxane networks
The kinetics of the surface gelation processes were measured by time-sweep-tests. In these experiments the evolution of the storage and loss moduli and, hence, cross-linking processes, could be observed as a function of time. The angular frequency ω and the strain amplitude γ remained constant during these measurements. The striking increase of µ' and µ'' points to the presence of viscoelastic networks. Typical results of these measurements are shown in figure 3.

Figure 3. Time-sweep-experiment showing the kinetics of surface gelation (OTS solution with Γ = 12 molecules/nm², c = 0.00274 mM, ω = 1 rad/s, γ = 0.2 %).

After two hours polymerisation time the gelation point (µ' = µ'') was reached and the storage modulus increased strongly up to values of 0.33 Nm⁻¹. The loss modulus remained nearly constant with values about one decade smaller. The membrane showed striking viscoelastic properties. When both moduli µ' and µ'' attained the plateau regime we supposed that the network formation was completed. In order to make sure that the capsules did not continue their polymerization after desired time scales, we stopped this chemical reaction by removing the water and transferring the capsules into anhydrous glycerol.

It turned out that many parameters had influence on the gelation process such as the OTS concentration and the mixture of the organic solvents. These processes were extensively investigated in our group [11, 23-25]. In this context it is interesting, to note, that the addition of 1,2,4-trichlorobenzene to p-xylene showed similar values of µ' in the range of Γ = 6 and 12 molecules per nm² as the pure p-xylene solution. On grounds of this special situation we performed the experiments at these conditions [23]. The elastic modulus slightly increased with rising OTS concentrations but it was not greatly influenced by the water content of the organic phase [11].

A typical frequency-sweep-test is presented in figure 4. In this experiment the angular frequency was varied between 0.01 and 100 rad/s and the strain amplitude was adjusted to γ = 0.2. The constant curve progression of µ' pointed to the existence of a chemical cross-linked, rubber-elastic membrane.
The relative large scattering of the loss moduli values was induced by the large differences between $\mu'$ and $\mu''$ and the bad signal to noise relation of the torque transducer for the evaluation of these experimental data.

The non-linear viscoelastic properties of the polysiloxane networks are represented in figure 5. The upper limit of the linear viscoelastic range (LVR) was determined from the descent of the elastic modulus. The synthesized membranes showed a rather small LVR of about 1%. We therefore applied small deformations below this yield value so that the membrane response could still be described by linear differential equations. Above the LVR we also observed irreversible deformations and network rupture.

3.2. Microcapsules in simple shear flow

3.2.1. Folding and bursting processes. In a series of experiments we investigated microcapsules with OTS concentrations of 0.2 and 0.4 mM at different polymerization times. In contrast to the time-sweep-time-tests (figure 3), network formation of the curved membranes occurred much faster. This could be determined by measurements in the optical flow cell regarding the deformation as a function of shear rate. In comparison to similar emulsion droplets the deformation curves of capsules showed lower slopes. This is easy to understand because instead of interfacial tension, the elasticity of the membrane creates resistance to deformation. Typical elastic moduli for polysiloxane capsules with $\Gamma = 6$ and 12 molecules/nm$^2$ range between $E_s = 1.47$ and 1.68 Nm$^{-1}$ (polymerization time of 60 min.) [11].

The surface Young’s moduli $E_s$ were calculated from the linear slope between the measured deformation $D$ and the shear rate $\dot{\gamma}$ according to equation 2.

Figure 6 presents a typical result of rheoscopic experiments, which illustrates the difference between an emulsion droplet and a capsule (axis ratio $B/L = 0.99$). For both suspensions the viscosity ratio was identical ($\lambda=0.001$). While the emulsion droplet elongated continuously with raising shear rate, the capsule deformed only slightly. At a critical shear rate of about 25-26 s$^{-1}$ however, the capsule deformation increased rapidly. At these conditions irreversible membrane deformation followed by network rupture occurred.
Close inspection revealed that the capsule showed wrinkling instabilities during flow. These folding processes always appeared parallel to the main axis of the capsule (figure 7). With increasing shear rate the folding was more pronounced, but reaching the membrane rupture point they became blurred and finally disappeared. After breaking the deformation of the capsule could easily be followed with the naked eye, because elongation proceeded rapidly. In this range we should rather talk about an emulsion droplet because the membrane was not intact anymore. It is evident that the slope of the broken capsule is steeper than the curve of the pure emulsion droplet. This phenomenon can be explained by membrane fragments remaining on the surface which lower the interfacial tension. It is worthwhile to mention that the disappearance of wrinkling instabilities was accompanied by rupture processes of the polysiloxane networks.

The appearance of folding processes is even more difficult to understand. They occurred when shear forces were acting on the surface of the capsules (figure 7). An explanation for this phenomenon might be the presence of negative membrane tensions. It was recently suggested that osmotic pressures could avoid folding processes by inducing positive membrane stresses [8]. Since the measurements were carried out under ambient air and at room temperature, we cannot exclude that tiny amounts of water were dissolved in the different phases. Especially glycerol is very hygroscopic. The water content of the organic solution should not be affected much because it was stored under argon and used immediately after preparing. In order to calculate the magnitude of osmotic pressures the solubility of water molecules and glycerol in the oil phase must be considered. As these solubilities were very low [11], the resulting pressure gradients can only be small. In a series of experiments we did not observe any volume changes of the capsules which point to the presence of transport processes across the interfaces. For these reasons the effects of osmotic pressures can be neglected.

It is also conceivable that wrinkling instabilities depend on the bending elasticity of the surrounding membranes. Minor changes during the surface gelation can lead to slightly deviations of membrane properties and might be responsible for the different behavior of capsules. It is also possible
that the membranes are pre-stressed and not in an equilibrium state. This can be caused during the synthesis by small shear stresses, gravitational forces or diffusion processes.

According to recent studies of Yazdani et al. [21] the wrinkles should disappear in the presence of membrane viscosity when the viscosity ratio $\lambda$ increased. In our case both the viscosity ratio ($\lambda = 0.001$) and the membrane viscosity ($\mu'$) were very small. It is, hence, difficult to estimate in which degree the membrane viscosity is involved in folding processes. At the present state it seems also possible that the bending resistance or the mechanical anisotropy, which is caused by oriented membrane molecules, induce buckling instabilities. Polysiloxane membranes very often showed this phenomenon. Membrane folding processes might even exist in the quiescent state if their sizes are below the limit of the optical resolution of our microscope. Such small phenomena are, of course, not visible to the naked human eye.

A series of very small membrane wrinkling processes are shown in figure 7 (red rectangle). During capsule movement these processes are generally easier to observe in a video film. However, the interesting aspect here is another phenomenon: the bursting process. As soon as the membrane breaks, the wrinkling processes disappear and the capsule deformation increases.

![Figure 8. Bursting and wrinkling process of a microcapsule in simple shear flow. (Polymerisation time = 94 min., $\Gamma = 12$ molecules/nm², $T = 26^\circ$C , $B/L = 0.97$).](image)

During the whole measurement the capsule deformation remained nearly constant until a critical shear rate of 42-43 s$^{-1}$ was reached (see also figure 9). The snapshot images at shear rates of 0 and 33 s$^{-1}$ illustrate that no remarkable capsule deformation occurred in this range. This small deformability agrees well with the results of the LVR.

![Figure 9. The deformation curve of an microcapsule in shear flow; polymerisation time = 94 min., $\Gamma = 12$ molecules/nm², $T = 26^\circ$C , $B/L = 0.97$.](image)

At $\dot{\gamma} = 42.5$ s$^{-1}$ the shear forces were sufficient to induce a bulge which initiated the bursting. The capsule membrane was destroyed within a few seconds. This means that the deformation proceeded so
fast that even the now generated emulsion droplet (figure 8 last photo) soon teared apart. The almost vertically slope in figure 9 gives an impression of this very rapid bursting mechanism.

3.2.2. Tumbling and swinging motions. Non-spherical particles showed an interesting orientation behavior. This process was very different from the performance of spherical capsules. As the viscosity ratio was adjusted to be constant ($\lambda = 0.001$), the shear rate was the only parameter which could be varied. In figure 9 different oscillation motions of a capsule with an axis ratio of $B/L = 0.96$ are presented. In regimes of small shear rates we observed tumbling motions. In this mode the inclination angle varied continuously between $+90^\circ$ to $-90^\circ$ (figure 10), which correlates to one half turn of the capsule rotation. Shape oscillations accompany this motion (figure 10). First inspections revealed that the capsule tumbled similar to a rigid body due to its small deformability. Obviously the hydrodynamic forces were not large enough to align the capsule in the flow direction. At evaluated shear rates, however, a swinging motion was detected (figure 11), superimposed by a tank-treading motion. Here, the inclination angle varied around positive values while the membrane rotated around its core. Tank-treading could be observed if tracer particles were used or inhomogeneities on the membrane surface were visible.

Figure 10. Tumbling motion of a microcapsule with an axis ratio of $B/L = 0.96$; polymerization time = 75 min., $\Gamma = 12$ molecules/nm², $T = 26^\circ$C.

Figure 11. Swinging motion of a microcapsule with an axis ratio of $B/L = 0.96$; polymerization time = 75 min., $\Gamma = 12$ molecules/nm², $T = 26^\circ$C.

Obviously the growth of shear forces induced TT and orientated the capsule to positive inclination angles. The oscillations around a constant value were caused by the membrane rotation and the “shape memory effect” [20] and therefore by periodic changes of the capsule shapes. The minor and major axis of the flexible particles varies continuously. It is worthwhile, to mention, that with rising shear rate the amplitude diminished and finally faded away. This oscillation decay was another indication for capsule bursting. It correlated very well with the steep rise of the deformation, and both phenomena occurred simultaneously (figure 12). The transition from TU to SW usually occurred via an intermittent motion. This regime was characterized by single oscillations ranging from $+80^\circ$ to $-80^\circ$, $+80^\circ$ to $-30^\circ$ and $+75^\circ$ to $-85^\circ$ (figure 13).

At these conditions the capsule failed to make a complete tumble. Due to the growing shear forces the capsule underwent shape oscillations in which a nearly spherical contour was reached. This globular shape only existed for an instant, but made it difficult to determine an axis and an inclination angle. It was evident that the different capsule shapes during the oscillation period caused these varying orientation fluctuations.

For the capsule presented in figure 13 the IR was slightly different from the motion described by Skotheim, Secomb and Sui et al. [1,20]. In this investigation the SW mode was only interrupted by
one single tumble motion. The inclination angles changed between positive and negative values, but did not reach -90° and +90°. For a quantitative comparison between theory and experiments it must be considered that the numerical simulations [1] were carried out with a viscosity ratio of $\lambda = 1$, and we have, instead, a very low value of $\lambda = 0.001$.

**Figure 12.** Swinging regime of a microcapsule with an axis ratio of $B/L = 0.98$; polymerization time = 70 min., $\Gamma$ = 6 molecules/nm², $T=25^\circ$C.

**Figure 13.** Intermittent and swinging regime of a microcapsule with an axis ratio of $B/L = 0.98$; polymerization time = 70 min., $\Gamma$ = 6 molecules/nm², $T=25^\circ$C.

4. Conclusion

In a series of experiments we systematically measured the shear induced deformation and orientation behaviour of polysiloxane microcapsules. These particles showed a rather small linear-viscoelastic regime up to deformations of about 1%. Above this yield value non-linear properties and membrane bursting occurred.

For a large number of capsules we observed shear induced membrane wrinkling instabilities. These folding processes were probably induced by pre-stressed membranes, the bending resistance or the mechanical anisotropy of the surrounding polymer layers. Network rupture immediately quenched the presence of wrinkling processes.

For non-spherical capsules we observed at least three different motions and periodic oscillations. In the regime of low shear rates tumbling motion occurred similar to the behaviour of a rigid body. At increasing shear forces we detected a swinging mode which was accompanied by a tank-treading membrane rotation. The transition from tumbling to swinging occurred via an intermittent regime.

References

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