Observations of Liquid-Liquid Encapsulation in Coextrusion of Inelastic Newtonian Fluids

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Abstract

The conditions under which liquid-liquid encapsulation can occur in coextrusion of polymers have been experimentally studied. The objective of this study was to determine if liquid-liquid encapsulation is a viscous phenomenon, viscoelastic phenomenon, or a combination of the two. The experimental observations discussed herein focus on the role of viscous stresses on encapsulation in the absence of viscoelastic behavior. Experiments with two inelastic Newtonian fluids flowing side-by-side in a transparent channel were conducted. Different combinations of glycerol, silicone oil, and motor oil were employed for the bi-layer flow experiments. Irrespective of the difference in the viscosity, no core-annular encapsulation was observed for bi-layer flows of glycerol and silicone oil. In two of the experiments with bi-layer flow of motor oil and silicone oil, motor oil encapsulated the silicone oil, even when the viscosity of motor oil was higher than the viscosity of the silicone oil. Since the flow behavior remained unaffected even with large variation in the viscosity of the fluids employed, it is concluded that other fluid properties besides viscosity, such as wettability of the fluid, may play an important role in reaching the core-annular encapsulation often observed in bi-layer flows. When gravity was acting perpendicular to the flow direction the difference in the density of the fluids also affected the layer arrangement.

Introduction

Liquid-liquid encapsulation is used to describe the tendency of a liquid to displace another liquid in a direction perpendicular to the flow direction, often along the channel walls. This can result in one liquid forming an annulus by completely wrapping around a core of another fluid. Encapsulation has been observed in several fluidic transport applications including the coextrusion of viscoelastic polymers [1] as well as the lubricated pipelining of crude oil and water, which are purely viscous in nature [2]. There is disagreement in the literature on whether viscosity difference alone result in encapsulation or if other fluid properties need to be considered as well. Some experimental observations report encapsulation is driven by viscosity differences, but no model has been shown to properly explain the behavior [3]. Multiple unsuccessful attempts have been made at numerical prediction of the encapsulation phenomenon using generalized Newtonian formulations [4-6]. This indicates that a fluid property other than viscosity difference may affect the initiation and development of encapsulation. The results of an experimental parametric study of bi-layer flow of several different fluid combinations are reported in this paper.

Experimental Setup

Transparent channels were constructed to observe the interaction of two fluids in a bi-layer flow. A 115 mm long 1 mm by 1 mm groove was micro-milled out of a smooth 12.5 cm thick polycarbonate plate such that two fluids enter the channel at separate ports and converge at an angle of 60° in a Y configuration. The machined surface of the plate was clamped to another polycarbonate plate to form the fourth wall of a square channel.

Transparent fluids were necessary in order to see through the entire depth of the channel. Liquid-liquid diffusion was avoided by running the experiments with immiscible fluid pairs. One fluid pair was glycerol (Sigma Aldrich) with polydimethylsiloxane (PDMS) silicone oil (Gelest, Inc.). Another fluid pair was motor oil (Valvoline) with the same

PDMS silicone oil. Four different viscosity grades of PDMS silicone oil were used with the glycerol and motor oil to experiment with different viscosity ratios between the fluids. All of these fluids are Newtonian with fluid properties listed in Table 1.

Viscosities of the fluids were determined using Cannon-Fenske viscometers. Specific gravity values in Table 1 are based on manufacturer specifications. The contact angle of glycerol was measured by placing a drop of the liquid onto a polycarbonate surface and observing the angle, θ, that the fluid formed at the solid-liquid-air interface. Figure 1 shows an illustration of this contact angle measurement. The contact angles for the motor oil and the silicone oils were too small to be accurately measured by viewing a liquid drop on a polycarbonate plate from the orientation indicated in Figure 1. Approximations of the contact angles for these fluids are shown in Table 1. Accurate measurements of the contact angle for motor oil and silicone oil are currently being performed.

Accurately visualizing the three-dimensional bi-layer flow required an ability to focus on different planes at various depths within the channel. To achieve a variabledepth focus, fluorescent microparticles were added to the

fluids. A fluorescent light source was used for excitation of the fluorescent microparticles and a stereomicroscope with fluorescent filters allowed the particles to be focused upon at different depths of the channel.

All experiments were performed at room temperature with syringe pumps moving the fluids through the channel at matched flow rates. The fluids were introduced into the channel by first filling the channel with one fluid, and then introducing the second fluid to generate a layered flow condition. All experiments were repeated by alternating which fluid was introduced to the channel first to observe any difference in the final flow configuration. The experiments were repeated with the channels oriented horizontally and vertically to verify that gravity was not inducing or hindering any encapsulation effects in any of the experiments. The fluids were pumped over a range of flow rates from 0.12 to 3.58 mm^3/sec. All of the reported observations are based on repeated results from multiple experiment trials.

Experimental Observations

The configuration of the two fluids at the initial contact with each other at the Y intersection is referred to as a side-by-side configuration and is illustrated in Figure 2. As the side-by-side co-flow fluids travelled down the channel the final flow configuration at the exit either (i) remained unchanged, (ii) remained side-by-side with the fluid interface oriented in a new direction with respect to gravity, or (iii) evolved to fully encapsulated flow as illustrated in Figure 3.

Values of Reynolds, capillary, and Bond numbers are listed in Table 2 to quantify the relations of inertia to viscosity, viscosity to surface tension, and gravity to surface tension observed in each experimental set. Approximate values of dimensionless numbers from coextrusion of polyethylene performed by Dooley and Rudolph [1] are included in Table 2 for comparison with the conditions and fluids used in these experiments. The Reynolds and Bond numbers indicate that inertia and gravity have small influence on the liquid-liquid systems as in a typical polymer coextrusion. However, the capillary numbers indicate that surface tension effects here are more prevalent relative to viscous effects here than in polymer coextrusion. Despite the capillary number differences, encapsulation as observed in polymer coextrusion has also been observed in some of the experiments in this work.

Glycerol and PDMS Silicone Oil

The glycerol-silicone oil experiments never exhibited encapsulation regardless of the viscosity difference between the two liquids. The initial side-by-side flow configuration was maintained throughout the length of the channel even though glycerol was much more viscous than the silicone oils A, B, C, and D as listed in Table 1.

In the set of experiments where gravity acted perpendicular to the flow direction, buoyancy-induced layer rearrangement was observed. The lighter silicone oil displayed a tendency to gravitationally stratify on top of the heavier glycerol while the large contact angle of the glycerol resisted deformation and limited the development of the buoyancy driven layer rearrangement. Additionally, the order in which the liquids were introduced affected the extent of the development of gravitational stratification relative to the initial side-by-side configuration. As the thinner and more wetting fluid, silicone oil did not displace the glycerol as easily as the glycerol displaced the silicone oil. Figures 4 and 5 illustrate a situation in which different final flow configurations were observed by changing only the order in which the fluids were introduced to the system.

Motor Oil and PDMS Silicone Oil

Encapsulation was observed for certain motor oilsilicone oil combinations. When the high viscosity silicone oil (A) was used, the less viscous motor oil fully encapsulated the more viscous silicone oil regardless of the orientation of gravity. Images of encapsulated flow from the motor oil-silicone oil (A) experiments are shown in Figures 6 and 7.

When silicone oil (B), which has a smaller viscosity than that of the motor oil, was used, the more viscous motor oil fully encapsulated the less viscous silicone oil regardless of the orientation of gravity. Figures 8 and 9 show the encapsulated flow from these experiments.

When either silicone oils (C) or (D) were used with motor oil and the gravity was along the flow direction, in spite of the large difference between the viscosities of the motor oil and the two silicone oils the initial side-by-side flow configuration was maintained for the length of the channel. The side-by-side flow configuration in the absence of buoyancy driven layer rearrangement is depicted in Figures 10 and 11. When the gravity acted perpendicular to the flow direction gravitational reorientation occurred in both of these sets of experiments. The buoyancy-induced layer rearrangement that occurred in these experiments is illustrated with Figures 12 and 13.

Discussion

Glycerol and PDMS Silicone Oil

The relatively large contact angle of glycerol was an important factor in the final flow configuration in these experiments. Figure 5 illustrates that the gravitational layer rearrangement never reached a fully stratified state in the glycerol-silicone oil experiments when gravity acted perpendicular to the flow direction. In contrast, Figures 12 and 13 illustrate that in the motor oil-silicone oil experiments the gravitational layer rearrangement always reached a fully stratified state. These differing observations can be explained by the large contact angle of the glycerol. The forces necessary to deform the glycerol contact line perpendicular to the flow direction had an apparent minimizing effect on the extent to which any gravitational layer rearrangement occurred. Subsequently, the less viscous silicone oil did not encapsulate the more viscous glycerol as expected. The absence of encapsulation may have been due to contact line pinning at the liquid-liquid-solid interface.

Motor Oil and PDMS Silicone Oil

Encapsulation as reported by the literature claims that the less viscous liquid will encapsulate the more viscous liquid [1-6]. With silicone oil (A) this encapsulation was observed. The experiments with silicone oil (B) and motor oil resulted in the more viscous liquid encapsulating the less viscous liquid. Because this "inverse encapsulation" occurred in purely viscous fluids, an analysis suggests that at least one property of Newtonian liquids other than viscosity plays a role in the onset of liquid-liquid encapsulation. The experiments with silicone oils (C) and (D) with motor oil resulted in no encapsulation. Just like with the glycerol experiments it is suggested that the larger contact angle of the motor oil could have been resisting the displacement by the thinner silicone oils along the channel walls.

Conclusions

Transparent Newtonian fluids pumped through a polycarbonate channel were visualized through a stereomicroscope via fluorescent excitation of microparticles. Either side-by-side or encapsulated flow were observed as the final flow configuration in all of the experimental combinations. Gravitational influence was observed in all experiments in which gravity was acting perpendicular to the direction of flow. However, the effect of gravity did not change the final flow configuration from the type which was observed when the gravity was acting parallel to the direction of flow.

Encapsulation did not occur between glycerol and any of the silicone oils. The relatively large contact angle of the glycerol is expected to have been nullifying any force imbalance that would otherwise have initiated encapsulation.

Encapsulation as described by the literature occurred between motor oil and silicone oil (A). In this situation the more viscous fluid was encapsulated by the less viscous fluid. However, the opposite type of encapsulation occurred between motor oil and silicone oil (B). Here the less viscous fluid was encapsulated by the more viscous fluid. There is currently no suggested explanation for this phenomenon. In the experiments of motor oil and silicone oils (C) and (D) a side-by-side configuration was the resultant flow configuration. The apparent contact angle difference between the fluids could once again lead to the explanation for the lack of encapsulation in these trials.

Acknowledgements

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References

1. J. Dooley and L. Rudolph, “Viscous and Elastic Effects in Polymer Coextrusion”, Journal of Plastic Film & Sheeting, 19, 111 (2003).
2. D. D. Joseph, “Lubricated pipelining”, Powder Technology, 94, 211 (1997).
3. P. Yue, C. Zhou, J. Dooley, and J. Feng, “Elastic encapsulation in bicomponent stratified flow of viscoelastic fluids”, Journal of Rheology, 52, 1027 (2008).
4. A. Karagiannis, A. N. Hyrmak, and J. Vlachopooulo, “Three-dimensional studies on bicomponent extrusion”, Rheologica Acta, 29, 71 (1990).
5. K. B. Sunwoo, S. J. Park, S. J. Lee, K. H. Ahn, and S. J. Lee, “Three-dimensional numerical simulation of nonisothermal coextrusion process with generalized Newtonian fluids.” Korea-Australia Rheology Journal, 12, 165 (2000).
6. M. Gupta, “Mesh Partitioning Technique for ThreeDimensional Simulation of Coextrusion”, SPE ANTEC Tech. Papers, 54, 217 (2008).

Keywords: Newtonian, encapsulation, contact angle, fluorescent.

Fluid	Viscosity (cP)	Specific Gravity	Contact Angle
Glycerol	934	1.262	79°
Motor Oil	112.6	0.858	< 30°
Silicone Oil A	191.4	0.968	< 20°
Silicone Oil B	46.6	0.96	< 20°
Silicone Oil C	9.2	0.935	< 5°
Silicone Oil D	4.2	0.918	< 5°

Table 1. Properties of fluids employed in experiments

Experimental Fluid Set	Reynolds No.	Capillary No.	Bond No.
Glycerol & Silicone Oil	5.8e-4 – 2.5e-3	8.2e-2 – 2.7e-1	9.0e-2 – 1.1e-1
Motor Oil & Silicone Oil A	8.1e-4 – 3.6e-3	1.5e-2 – 6.2e-2	3.4e-2
Motor Oil & Silicone Oil B	3.4e-3 – 9.8e-3	1.8e-2 – 3.5e-2	3.2e-2
Motor Oil & Silicone Oils C &D	3.1e-3 – 5.4e-1	2.4e-3 – 3.1e-1	1.8e-2 – 2.7e-2
Polyethylene*	4.7e-5 – 4.4e-4	2.6e+3 – 9.3e+3	0

*Calculated from experiments done by Dooley and Rudolph [1].

Table 2. Dimensional analysis of experiments

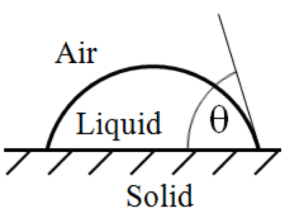

Figure 1. Depiction of solid-liquid-air contact angle measurement θ.

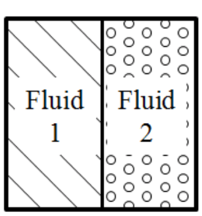

Figure 2. Side-by-side flow configuration.

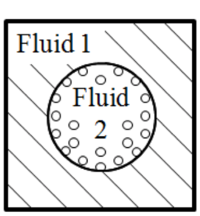

Figure 3. Fully encapsulated flow configuration. Fluid 1 forms an annulus and Fluid 2 forms a core.

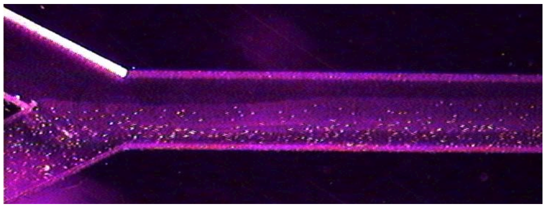

Figure 4. Silicone oil (A) (bottom) and glycerol (top) extruded left to right through the converging channel. Microparticles in the silicone oil are highlighted with the fluorescent light. Here gravity is oriented perpendicular to the page and buoyancy- induced stratification was observed. In this image buoyancy has altered the initial side-by-side configuration by moving one liquid-liquid-solid interface line up the page and the other down the page. The cross-sectional view shown in Figure 5(b) illustrates the final flow configuration of this image.

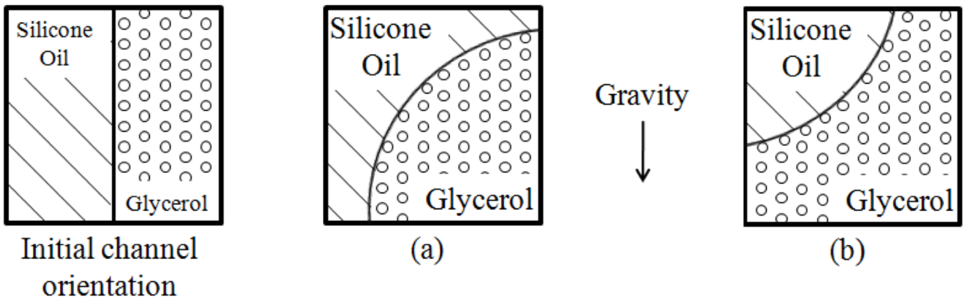

Figure 5. The final flow configuration of glycerol and silicone oil depicted in (a) resulted when the silicone oil was the first fluid in the channel and was displaced by glycerol. Provided that the exact same initial orientation was repeated, the flow configuration in (b) resulted when the glycerol was the first fluid in the channel and was displaced by silicone oil.

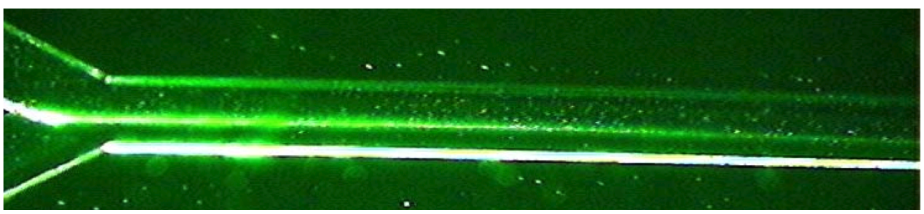

Figure 6. Silicone oil (A) (bottom) and motor oil (top) extruded left to right in the converging channel. Microparticles in the motor oil are highlighted with the fluorescent light filter. Here encapsulation was observed regardless of the orientation of gravity. The liquid-liquid-solid interface line seen in this image approached the side of the channel as the motor oil encapsulated the silicone oil.

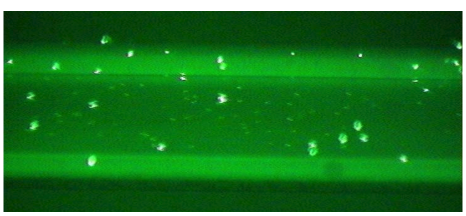

Figure 7. Silicone oil (A) (core- dark) and motor oil (annulus- light) extruded left to right. This magnification shows the fully encapsulated flow configuration with the silicone oil completely separated from all channel walls. Here the microparticles in the silicone oil are dimly visible while the microparticles in the motor oil are highlighted with the fluorescent lighting. In this image some of the microparticles in the motor oil are in focus and others are out of focus. This is an indication that the motor oil had fully encapsulated the silicone oil.

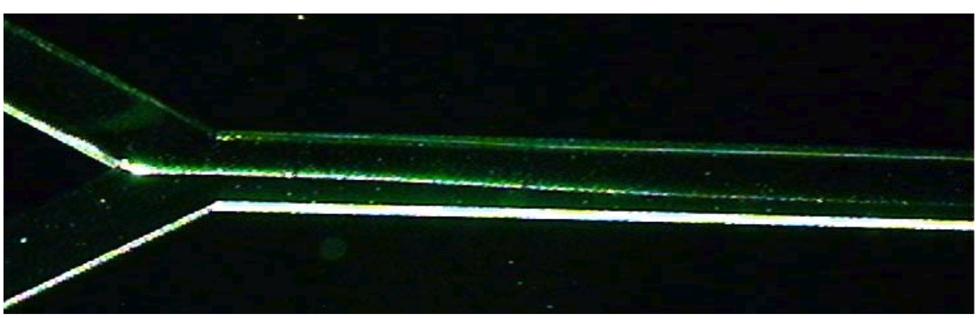

Figure 8. Silicone oil (B) (bottom) and motor oil (top) extruded left to right through the converging channel. Here

encapsulation was observed regardless of the orientation of gravity. The liquid-liquid-solid interface line seen in this image approached the side of the channel as the motor oil encapsulated the silicone oil.

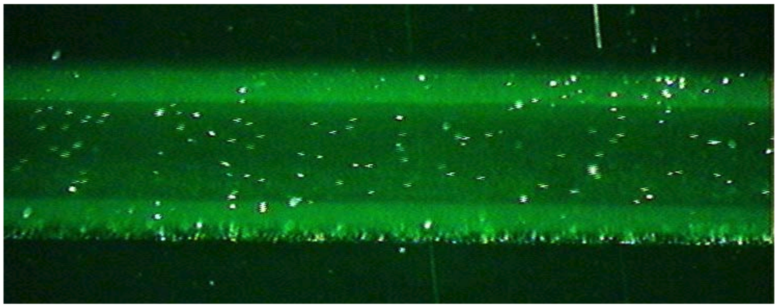

Figure 9. Silicone oil (B) (core- dark) and motor oil (annulus- light) extruded left to right. This magnification shows the fully encapsulated flow configuration with the silicone oil completely separated from all channel walls. Here the microparticles in the silicone oil are dimly visible while the microparticles in the motor oil are highlighted with the fluorescent lighting. In this image some of the microparticles in the motor oil are in focus and others are out of focus. This is an indication that the motor oil had fully encapsulated the silicone oil.

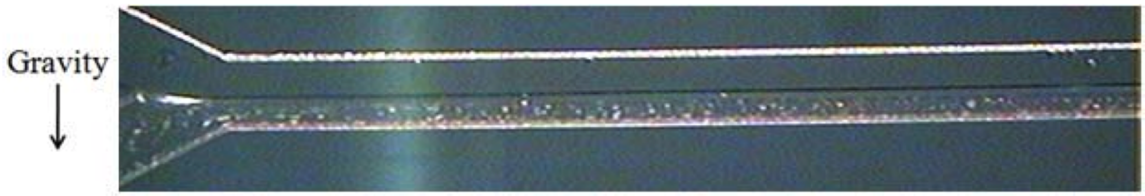

Figure 10. Silicone oil (D) (bottom) and motor oil (top) extruded left to right in the converging channel. Here buoyancy- induced layer rearrangement was not observed as the heavier silicone oil entered the channel already beneath the motor oil. The side-by-side flow configuration is shown here under normal lighting conditions.

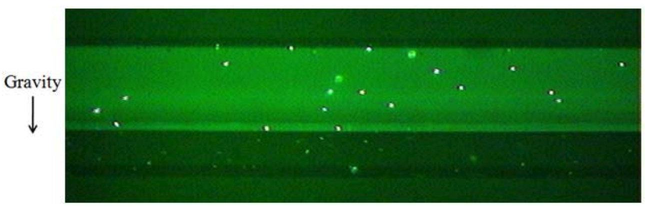

Figure 11. Silicone oil (C) (bottom) and motor oil (top) extruded left to right in the converging channel. Here buoyancy- induced layer rearrangement was not observed as the heavier silicone oil entered the channel already beneath the motor oil. A magnification of the side-by-side flow configuration is shown here with fluorescent lighting highlighting the microparticles in the motor oil.

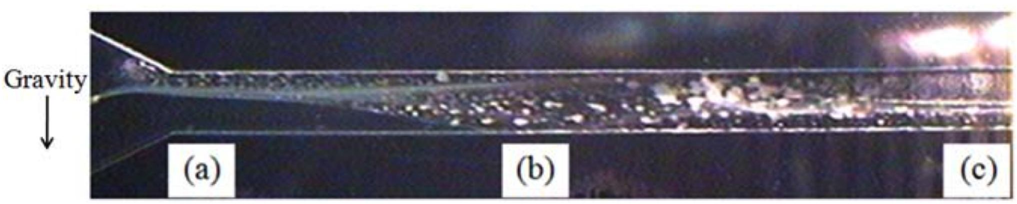

Figure 12. Silicone oil (C) (top at the intersection) and motor oil (bottom at the intersection) extruded left to right in the converging channel. Here buoyancy-induced layer rearrangement was observed as the heavier silicone oil did not enter the channel in a gravitationally stable arrangement with respect to the lighter motor oil. In the center of this figure the side-by- side configuration has rotated 90° relative to its original orientation. At the right edge of this figure the side-by-side flow configuration has rotated 180° relative to its original orientation. Cross-sectional images in Figure 13 illustrate the flow configuration at points (a), (b), and (c) in this image.

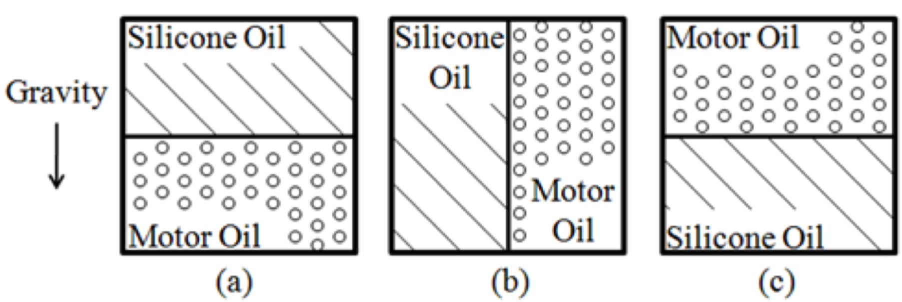

Figure 13. This sequence of cross-sectional images illustrates the gravitational layer rearrangement depicted in Figure 12.

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