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Interfacial instability of Boger fluid in the pull-off test between two circular plates

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Abstract

In this study the deformation of liquid-air interface of Newtonian or Boger fluids filled between two parallel-plates geometry was investigated when two surfaces were separated at a constant speed. The interface between the fluid and air showed either stable or unstable deformation depending on experimental conditions. Repeated experiments for a wide range of experimental conditions revealed that the deformation mode could be classified into three types: 'stable region', 'fingering' and 'cavitation'. The experimental condition for the mode of deformation was plotted in a capillary number vs. Deborah number phase plane. It has been found that the elasticity of Boger fluids destabilize the interface deformation. On the other hand, the elasticity suppresses the formation and growth of cavities.

Keywords **:** pull-off test, tack, pressure sensitive adhesive, viscous fingering, cavitation, Boger fluid

1. Introduction

Pull-off test is one of the tests for determining the bond strength of adhesives. It measures the force or work required to pull apart two surfaces separated by a thin film of adhesive. In the pull-off test, the force is usually measured tack which enables to form a bond of measurable strength immediately on contact with another surface (Satas, 1999; Skeist, 1990). During the pull-off test, if stress exceeds a critical point, viscous fingering or cavitation is observed (Tirumkudulu and Russel, 2003). The presence of such phenomena is determined not only by the testing conditions such as the geometry of experiment, pull-of speed and initial gap size of test but also by the rheological properties of materials such as viscosity and elasticity. The complex nature of the rheology of real pressure sensitive adhesives (PSA) makes the analysis of tack very complicated. Real PSA usually displays elasticity and shear thinning simultaneously, which makes it more difficult to analyze the fingering and cavitation phenomena systematically. Recently Tirumkuludu and Russel (2003) and Tirumkuludu *et al.* (2003) have studied the tack and the interface deformation for Newtonian and shear thinning liquids. In the present study, the effect of elasticity on instability characteristics in the pull-off test was investigated by using polybutene based Boger fluids with a wide range of viscosity and elasticity to approach tack from a fundamental point of view. It has been found that the change in the shape of inner or exterior surfaces could be classified into three types, 'stable region', 'fingering' and 'cavitation'. It has been also found that elasticity destabilizes the interface of fluids. On the other hand, the elasticity appears to suppress the formation and growth of cavities.

2. Viscous fingering and cavitation

When a less dense fluid pushes denser fluid immiscible with the less dense fluid between two parallel plates (called Hele-Shaw cell) while keeping an immiscible interface under the influence of pressure gradient, the interface is subject to an unstable mechanism to show a wavy interface (Saffman and Taylor, 1958). As the wavy interface moves, the amplitude of the wave further grows and the wavy structure develops into long, fingerlike structure. This instability is called fingering (Homsy, 1987). In the case of Newtonian fluid, this instability is known to be dependent on capillary number defined as follows (Batchelor *at al.*, 2000):

$$Ca = \frac{U\mu}{\sigma}, \tag{1}$$

where U is the velocity of interface, μ is the viscosity of fluid and σ is the surface tension between two fluids. In the case of elastic fluids, Deborah number, De, should be also relevant, where De is defined as follows (Vlad and Maher, 2000):

$$De = \frac{\lambda}{T}, \tag{2}$$

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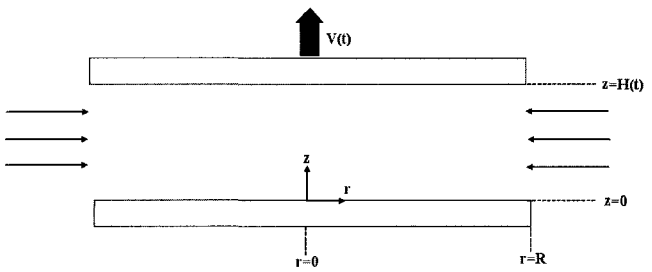

Fig. 1. Schematic diagram of the pull-off test.

where λ and T are material and processing times, respectively. In the pull-off tests using two parallel disks as shown schematically in Fig. 1, the air (less dense) pushes the liquid (denser) that fills the gap initially so that we expect to have fingering instability. Since the gap distance becomes larger as the pull-off motion continues, the velocity of air-liquid interface becomes smaller and the capillary number becomes larger, and hence the system could remain stable unlikely the case of Hele-Shaw cell. Therefore depending upon the pull-off speed, the interface could remain smooth without waves during the finite time span of the pull-off test.

When two surfaces are pulled off, the pressure distribution and the force required to keep the speed of motion are given as follows (Denn, 1980):

$$P(r,z) = p(r,z) - p_0 = \frac{3\eta V}{H}\left[2\frac{z}{H}\left(1-\frac{z}{H}\right)+\frac{r^2-R^2}{H^2}\right] \tag{3}$$

$$F = \frac{3\pi\eta V(t)R^2}{2H^3} \tag{4}$$

In the above equation, P is gauge pressure, p is the absolute pressure and $V(t)$ is pull-off speed. If the pull-off speed is sufficiently fast, due to $-R^2$ term in Eqn. (3), the pressure at the center falls off below the boiling point of the solution, which should be impossible physically. In this case a cavity is formed at center and grows as the pull-off motion begins. The presence of a cavity inside the fluid is called cavitation. As the pull-off motion continues, the cavity grows and then the interface between the cavity and the fluid is further subjected to fingering instability.

It is obvious that the presence of the fingering instability or cavitation affects the force required to pull off one of the plates from the other. This means that when fingering instability or cavitation occurs, the tack of pressure sensitive adhesives becomes different from the case without such phenomena. Hence it is very important to understand the flow pattern during pull-off tests depending on both physicochemical and rheological properties and the mode of pull-off motions. In this research we have investigated the effect of rheological properties and/or the pull off modes.

3. Experiment

3.1. Materials

As Newtonian fluids, polybutene (PB) with differing molecular weights were used. As the viscoelastic liquid, PB based Boger fluid was prepared to separate the effect of elasticity from the shear-thinning properties of polymeric liquids. The PB was purchased from Aldrich Chem. Co. and used as received. The molecular weights of PB were 320 and 920 Daltons (LPB and HPB, respectively). A mixture of PB with the molecular weight of 620 Daltons (MPB) was also prepared by mixing two fluids. PIB was supplied from BASF and also used as received. The PIB had the number average molecular weights of 250 and 425 kDaltons (LPIB and MPIB, respectively). Another PIB with the number average molecular weight of 3,100 kDaltons (HPIB) was purchased from Aldrich Chem. Co. and used as received.

It is very difficult to dissolve polymer with a high molecular weight in a very viscous solvent. To overcome this difficulty we followed the recipes used by Vermant *et al.* (2004) in which 2-chloropropane is used as the intermediate solvent for distributing PIB in PB. To remove the 2-

Table 1. Naming convention and compositions of Boger fluids

Component	Weight%	Nominal molecular weight
Solution MPIB_LPB		
Polyisobutylene	0.1, 0.3	425,000
2-chloropropane	<1	
Polybutene	>99	320
Solution HPIB_LPB		
Polyisobutylene	0.1, 0.3	3,100,000
2-chloropropane	<1	
Polybutene	>99	320
Solution MPIB_MPB		
Polyisobutylene	0.1, 0.3	425,000
2-chloropropane	<1	
Polybutene	>99	620
Solution HPIB_MPB		
Polyisobutylene	0.1, 0.3	3,100,000
2-chloropropane	<1	
Polybutene	>99	620
Solution LPIB_HPB		
Polyisobutylene	0.1, 0.3	250,000
2-chloropropane	<1	
Polybutene	>99	920
Solution MPIB_HPB		
Polyisobutylene	0.1, 0.3	425,000
2-chloropropane	<1	
Polybutene	>99	920

chloropropane for the mixture, a vacuum rotary evaporator was used. The final mixture was found to be free of microbubbles the diameter of which is larger than 1 μm when inspected under an optical microscope. The solutions prepared in this study is listed in Table 1.

3.2. Pull-off tests

Tack tests were performed by using a rotational rheometer (AR2000 Model, TA Instruments Co.) with a parallel-plates geometry (8 mm diameter, stainless steel). To observe the flow phenomena directly a homemade visualization apparatus was used as shown in Fig. 2. The lower plate of the apparatus was made of ground quartz not to be eroded chemically by PB or 2-chloropropane. Below the lower plate a 45° inclined mirror was installed. The visualization apparatus was set over the peltier of the rheometer. A digital camcorder (SONY DCR-VX2000) was used to record the time change of the air-liquid interface. It is important to maintain the alignment of the fixture since the minimum gap distance is as small as 20 micron. To achieve this we used an alignment tool to keep the maximum error in gap distance less than 1 μm. Since the wetting ability of the testing fluid used in this study is better than conventional PSAs, the contacting time effect was thought to be negligible in loading samples. To reduce the effect of preshear history of elastic materials, we waited 30 minutes after loading the sample before starting a pull-off experiment. The initial gap varied from 20 to 50 μm and the pull-off speed varied from 10 to 1000 μms^{-1}.

Even though the original purpose of this study was to understand pull-off tests with regard to the tack of pressure sensitive adhesives, it was impossible to measure the tack with the experimental apparatus used in this study. This is because the duration of each run of the pull-off test is about 2 seconds while during 2 seconds the rheometer can take

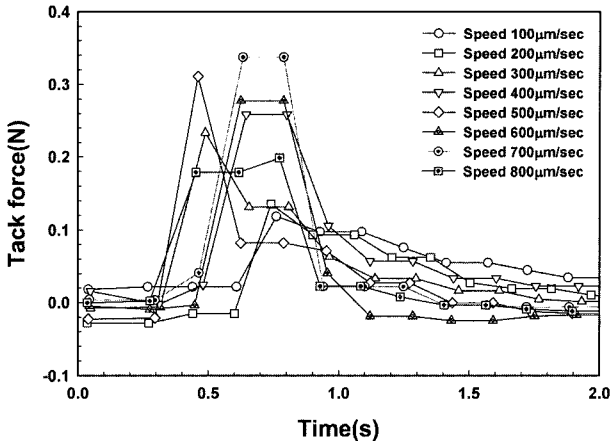

Fig. 3. The tack test result. Due to instrumental limit, tack force was not read properly. In this case, the tack force should be a monotonically increasing function of pull-off speed.

only 6 – 10 data points as shown in Fig. 3. This results in an abnormal value in the maximum force as shown in the figure. For example, when the pull-off speed exceeded 500 μms^{-1}, the maximum value decreased with pull-off speed. Therefore in the present study we focus on the interface pattern during the debonding process without considering the force.

3.3. Characterization of test fluid

In Fig. 4, the viscosity is plotted against shear rate for solutions listed in Table 1. The fluid exhibits the typical behavior of Boger Fluid: For three decades of shear rate, the viscosity shows almost no shear thinning and the change in viscosity does not exceed more than 20% in the experimental ranges considered here. The abrupt increases in viscosity when shear rate becomes larger than 100 s^{-1} for

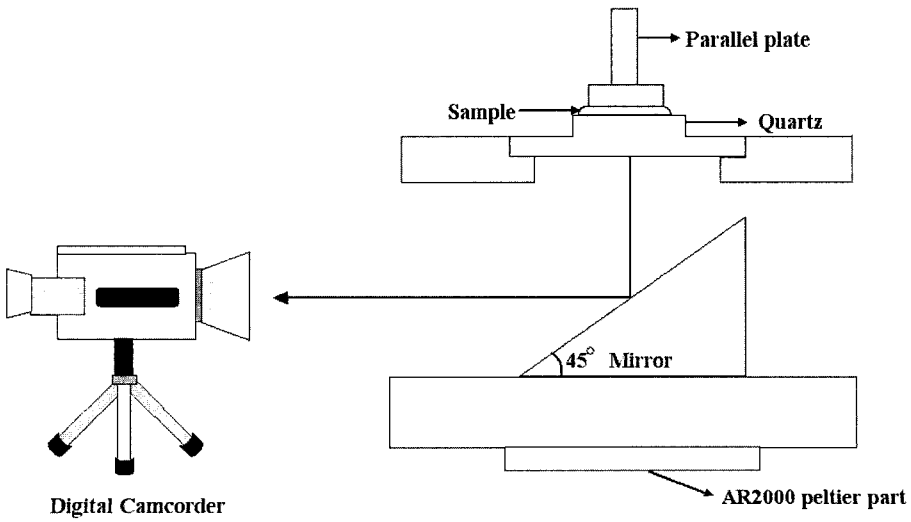

Fig. 2. Schematic diagram of the flow visualization apparatus.

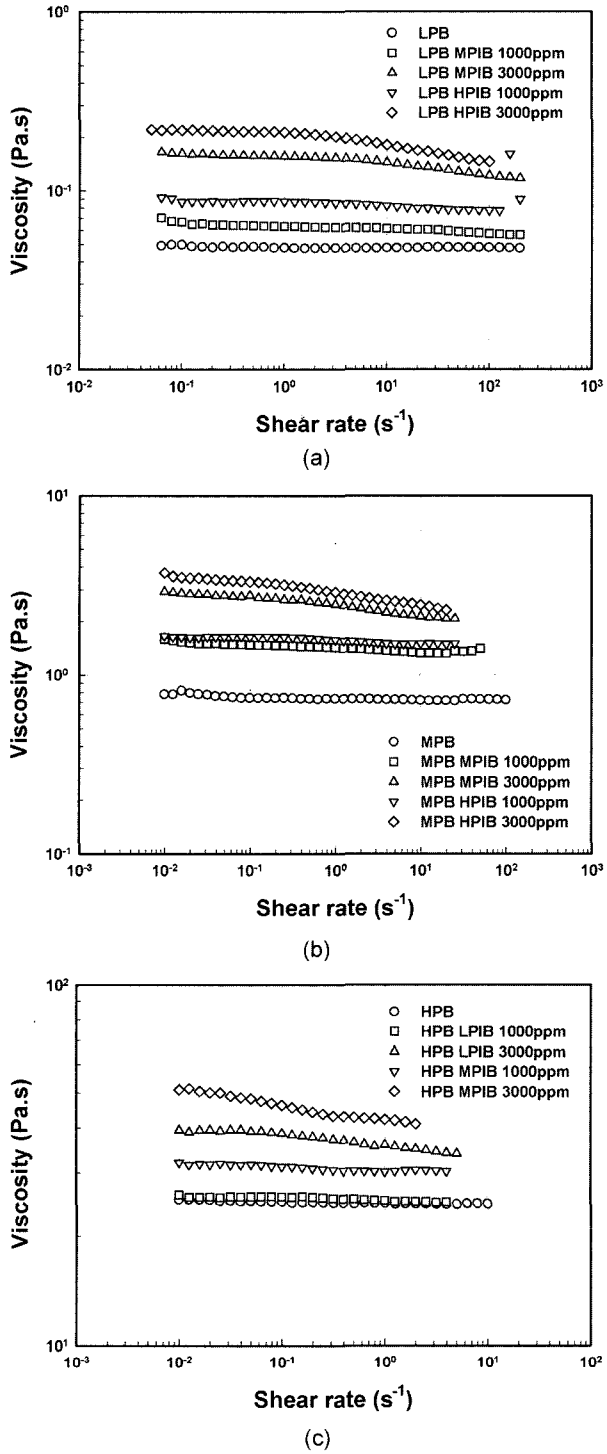

Fig. 4. Viscosity of Boger fluids. (a) LPB (MW of polybutene is 320); (b) MPB (MW polybutene is 620); (c) HPB (MW of polybutene is 920).

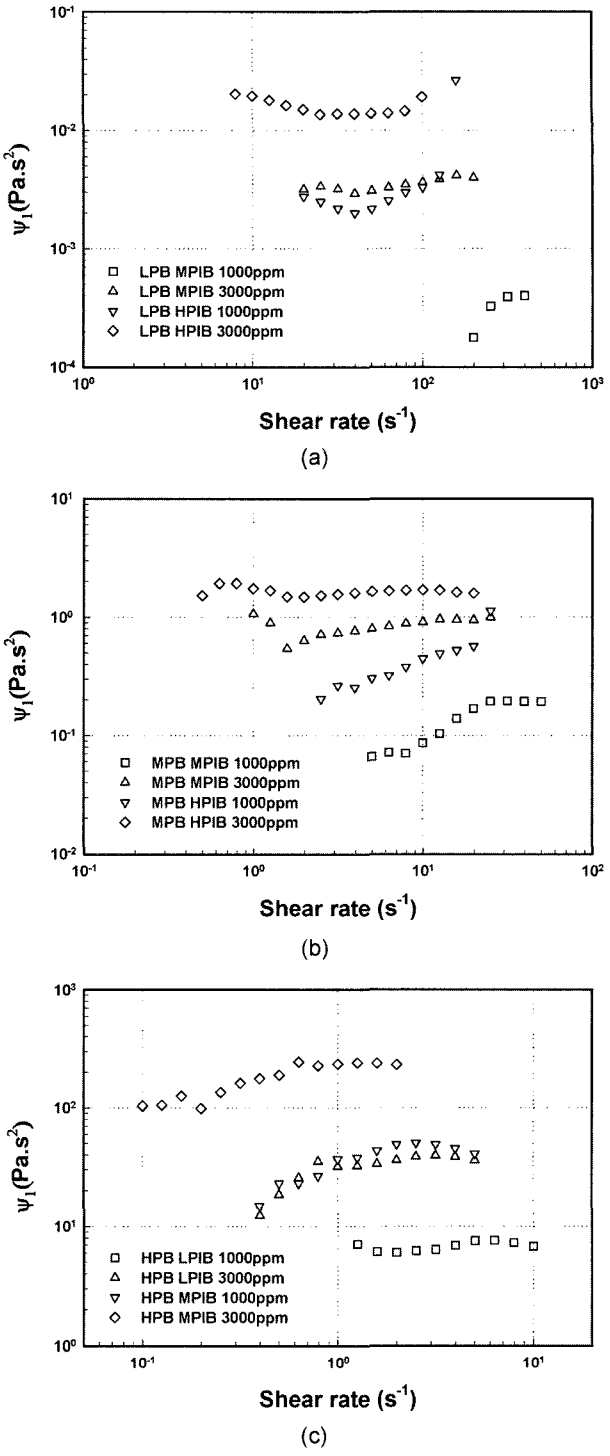

Fig. 5. The first normal stress difference coefficient of Boger fluids. (a) LPB (MW of polybutene is 320); (b) MPB (MW polybutene is 620); (c) HPB (MW of polybutene is 920).

LPB solutions in Fig. 4 appear to be due to elastic instability of Boger fluid. In Fig. 3, the first normal stress difference coefficient (Ψ_1) are plotted. Due to the instrumental limit, N_1 was measured only in the shear rate ranges shown in Fig. 5. For the shear rate range, Ψ_1 is approximately constant except for some cases. But we have to be careful whether this constant value is truly the zero-shear first normal stress difference coefficient (Ψ_{10})

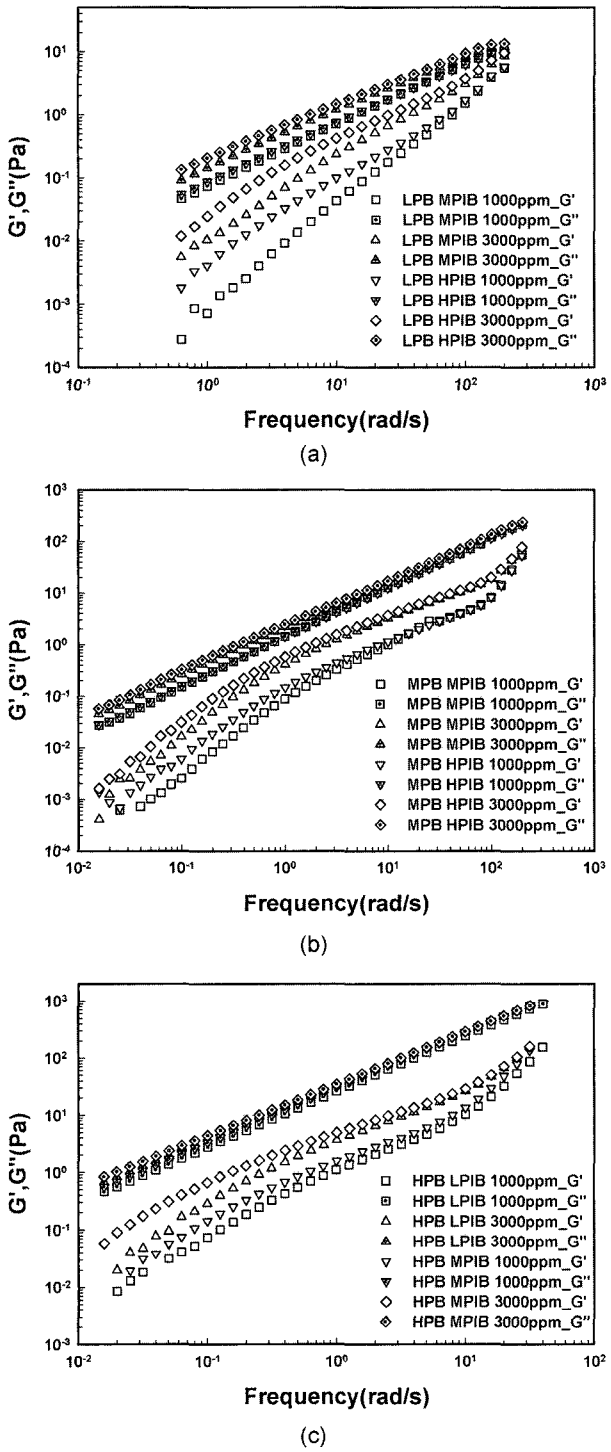

Fig. 6. Linear viscoelastic properties of Boger fluids. (a) LPB (MW of polybutene is 320); (b) MPB (MW polybutene is 620); (c) HPB (MW of polybutene is 920).

since Boger fluids could have the 2^{nd} plateau in Ψ_1 (Shaqfeh, 1996). Therefore in determining the relaxation time of Boger fluids, we did not use the normal stress difference data.

To determine the relaxation time of Boger fluid, we performed linear viscoelastic measurements as shown in Fig. 6. In all the cases, as the frequency tends to zero, the slopes of G' and G'' tend to 2 and 1 and the shapes of the curves show the typical characteristics of dilute polymer solution. Since it is well known that Boger fluid can be represented by Oldroyd-B model, we use the method suggested by Tenijenhuis (1990) as follows:

$$G' = \frac{\eta^2\omega^2(\lambda_1-\lambda_2)}{(1+\omega^2\lambda_1^2)} \tag{5}$$

$$G'' = \frac{\eta\omega(1+\omega^2\lambda_1\lambda_2)}{(1+\omega^2\lambda_1^2)} \tag{6}$$

$$\lim_{\omega\to 0}\frac{G'}{\omega^2} = \eta_0(\lambda_1-\lambda_2) \tag{7}$$

$$\lim_{\omega\to 0}\frac{G''}{\omega} = \eta_0 \tag{8}$$

$$\lim_{\omega\to\infty}\frac{G''}{\omega} = \eta_0\frac{\lambda_2}{\lambda_1} \tag{9}$$

where λ_1 and λ_2 are relaxation time and retardation time, respectively. The relaxation times were found to be in the range of 0.04 to 0.5 for LPB, 0.8 to 1.8 for HPB and 2 to 8 for HPB. The detailed values are shown in Table 2.

4. Results and discussion

In this study, we investigated the deformation of air-liquid interface of a liquid layer between two parallel disks when the upper disk is pulled off assuming a situation of measuring tack of PSA. After a sample is loaded between two plates, the upper plate is pulled off at a constant speed and the deformation of air-liquid interface was recorded with a camcorder. After testing for many differing sets of experimental conditions by varying materials, initial gap size and pull off speeds, we have been able to classify the debonding pattern into three different modes: stable and symmetrical shrink; fingering; cavitation followed by fingering.

In Fig. 7, a typical case of stable and symmetrical shrink patterns with time is shown. As the upper plate is pulled off, the fluid boundary advances while the radius becoming smaller and the fluid deforms while keeping almost the shape of a cylinder. In other words, the diameter at the center of the cylinder is almost the same as the diameter of the circular contact line touching the two solid surfaces. This behavior is observed when the viscosity of fluid is small and the pull-off speed is small. The second case of fingering is shown in Fig. 8. As the upper plate is pulled off the interface becomes wavy at first and the wave grows into large, finger-shaped boundary. Some of the fingers grow faster than the others and the large fingers suppress

Table 2. The rheological properties and surface tension of Newtonian solvent and Boger fluids.

Component	Viscosity (Pa·s)	Ψ_1 (Pa·s^2)	Relaxation time (s)	Density (g/cm^3)	Surface tension (mN/m)
Solvent LPB	0.05	-	0.	0.84	25.5
1000 ppm MPIB LPB	0.07	0.0004	0.038	0.84	26.6
3000 ppm MPIB LPB	0.16	0.004	0.125	0.84	29.0
1000 ppm HPIB LPB	0.09	0.004	0.088	0.84	27.6
3000 ppm HPIB LPB	0.22	0.015	0.468	0.84	29.0
Solvent MPB	0.78	-	0	0.865	29.4
1000 ppm MPIB MPB	1.58	0.15	0.711	0.865	30.1
3000 ppm MPIB MPB	2.92	0.80	0.946	0.865	30.2
1000 ppm HPIB MPB	1.65	0.50	0.755	0.865	27.5
3000 ppm HPIB MPB	3.55	1.60	1.904	0.865	30.3
Solvent HPB	25.24	-	0	0.89	32.9
1000 ppm LPIB HPB	26.05	7	2.040	0.89	33.6
3000 ppm LPIB HPB	39.44	35	3.286	0.89	34.2
1000 ppm MPIB HPB	32.02	45	4.503	0.89	34.1
3000 ppm MPIB HPB	51.77	220	8.240	0.89	34.6

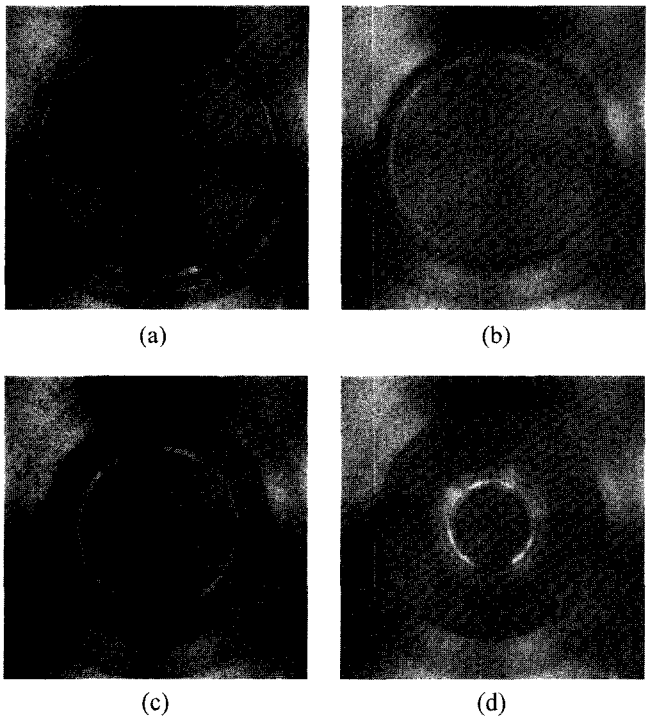

Fig. 7. Typical time evolution pictures showing a stable deformation of the air-liquid interface. The liquid is Newtonian polybutene (MW=620). (a) 0.33 s; (b) 1.00 s; (c) 2.00 s; (d) 6.00 s.

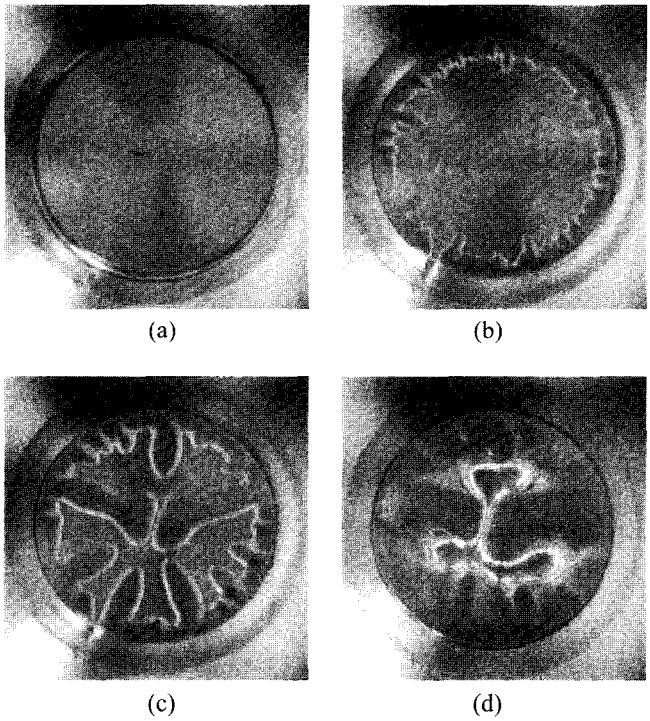

Fig. 8. Typical time evolution pictures showing fingering instability at the air-liquid interface. The liquid is Newtonian polybutene (MW=620). (a) 0.33 s; (b) 1.00 s; (c) 2.00 s; (d) 6.00 s.

the growth of small fingers to result in having only a few large fingers. The fingering is observed when the viscosity of fluid is larger and the pull-off speed is larger than the first case. The third case shows the formation of cavitation bubble at the center and the bubble grows as the pull-off motion continues (See Fig. 9). In this case the bubble-liquid interface becomes unstable to result in fingering instability. The process is very fast and the fingering pattern is different from the previous case in that tree like, fractal pattern is observed. But as the pull off continues, the cavity

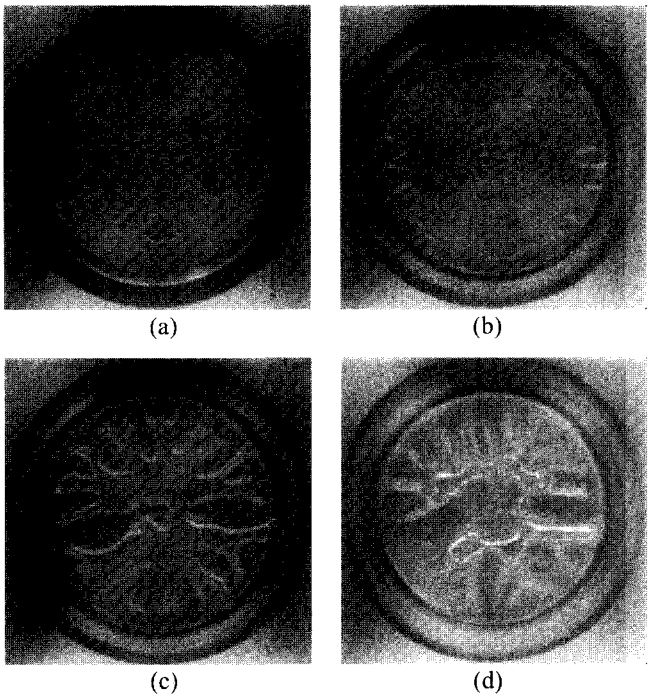

Fig. 9. Typical time evolution pictures showing the cavitation at the center followed by the fingering instability at the air-liquid interface. The liquid is Newtonian polybutene (MW=620). (a) 0.33 s; (b) 1.00 s; (c) 2.00 s; (d) 6.00 s.

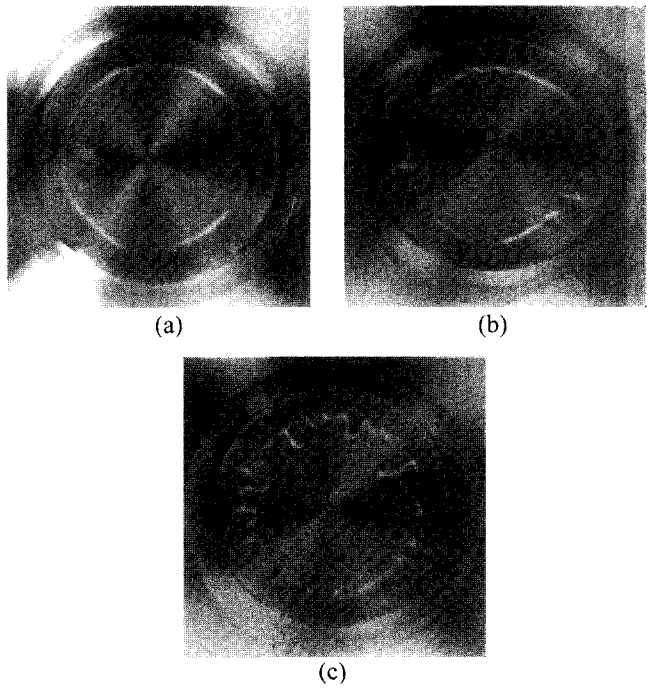

Fig. 10. Comparison of instability in the same viscosity sample (Initial gap: 50 μm, pull-off speed: 1000 μms^{-1}). (a) Viscosity: 4 Pa·s, surface tension: 30 mN/m, relaxation time: 0 s; (b) Viscosity: 2.92 Pa·s, surface tension: 30.2 mN/m, relaxation time: 1 s; (c) Viscosity: 3.55 Pa·s, surface tension: 30.3 mN/m, relaxation time: 1.9 s.

collapses and the outer boundary begins to form fingers. The cavitation occurs when the pull-off speed is sufficiently large that the pressure at the center becomes negative. The same three different modes were also observed when Boger fluids were tested.

One of the most important issues in the rheology of pressure sensitive adhesives is the effect of elasticity. To examine the effect of elasticity we have chosen three different kinds of fluids with almost the same viscosity with differing elasticity so that the capillary number for each run is almost the same while Deborah number is different: one Newtonian fluid and two kinds of Boger fluid with different relaxation times. In Fig. 10, the debonding characteristics for three different fluids are compared. In the figure, the interface becomes wavier as relaxation time becomes larger. This means that the elasticity of fluid destabilize the interface during the debonding process.

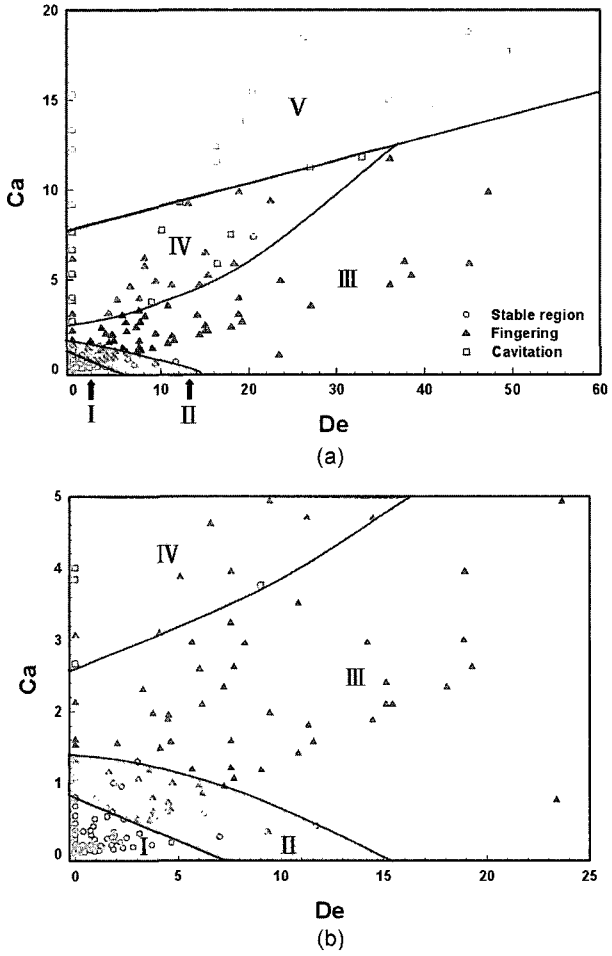

Fig. 11. Phase diagram of the different types of debonding phenomena as a function of *De* and *Ca*. (b) is the magnifying diagram of the lower left corner of (a). I: stable region, II: stable or fingering region, III: fingering region, IV: fingering or cavitation region, V: cavitation region.

Vlad and Maher (2000) obtained a similar destabilizing effect of elasticity in the case of Hele-Shaw flow of Boger fluids. The flow between two parallel plates during debonding is basically a radial flow and it is mostly elongational in nature. Therefore the instability is not caused by the hoop stress of elastic fluid. More study is needed to understand the destabilizing mechanism of elastic stress.

In Fig. 11, phase diagrams are shown dividing the debonding mode depending on *Ca* and *De*. In region I, the interface is stable during the whole debonding process. In region II, stable deformation of the interface and viscous fingering coexist. Whether there will be viscous fingering or not in region II depends on the slightly different experimental (including initial and/or fluid) conditions that could not be controlled virtually during the experiment. The region III represents the viscous fingering instability. It is noted that the slopes of the boundaries dividing regions I, II and III are negative. This means that the elastic properties of Boger fluid destabilize the interface and hence the fingering occurs at smaller *Ca* with increasing *De* as the interface deforms during the pull-off tests. The region V represents the condition where cavitation occurs while the region IV represents the coexistence of fingering and cavitation depending upon experimental condition. Unlikely the case of the slope of the boundaries dividing region I, II and III, the slopes of the boundaries dividing regions II, IV and V are positive indicating that the elastic property of Boger fluid suppress the formation of cavity at the center. The suppression of cavitation by elasticity has been also reported in the study of cavitaton bubbles in drag reducing solutions (Ellis *et al.*, 1970).

5. Conclusions

In this study the deformation of liquid-air interface of Newtonian or Boger fluids filled between two parallel-plates geometry was investigated when two surfaces were separated at a constant speed considering a situation of measuring tack of pressure sensitive adhesive. The Boger fluids were used to avoid the complexity of shear-thinning effect. The Boger fluid was prepared by dissolving polyisobutylene (PIB) in polybutene (PB). The concentration and number average molecular weight of PIB and PB were varied to control the viscosity and elasticity of Boger fluid. Using a commercial rheometer and homemade visualization apparatus, the deformation of air-liquid interface was examined. It has been found that the interface between the fluid and air shows either stable or unstable deformation depending on experimental conditions. Repeated experiments for a wide range of experimental conditions revealed that the deformation mode could be classified into three types: 'stable region', 'fingering' and 'cavitation'. The experimental condition for the mode of deformation was plotted in a capillary number vs. Deborah number phase plane. It has been found that the elasticity of Boger fluids destabilize the interface deformation. On the other hand, the elasticity suppresses the formation and growth of cavities. Due to the instrumental limit, it was not possible to correlate the interface deformation with tack force. Further studies are also needed on the tack force and interface deformation for shear thinning elastic liquids.

Acknowledgment

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References

Batchelor, G.K., H.K. Moffat and M. G. Worster, 2000, Perspectives in fluid dynamics, Cambridge Univ. Press, Cambridge.

Denn, M.M., 1980, Process Fluid Mechanics, Prentice-Hall Inc., New Jersey.

Ellis, A.T., J.G. Waugh and R.Y. Ting, 1970, *Trans. ASME J. Basic Eng.* **92**, 459.

Homsy, G.M., 1987, Viscous Fingering in Porous Media, *Annu. Rev. of Fluid Mech.* **19**, 271.

Saffman, P.G. and G.I. Taylor, 1958, The penetration of a fluid into a porous medium of Hele-Shaw cell containing a more viscous liquid, *Proc. Roy. Soc. London A* **245**, 312.

Satas, D., 1999, Handbook of Pressure Sensitive Adhesive Technology, 3rd ed., Satas & Associates, Warwick.

Shaqfeh, E.S.G., 1996, Purely elastic instabilities in viscometric flows, *Annu. Rev. Fluid Mech.* **28**, 129.

Skeist, I, 1990, Handbook of adhesives, 3rd ed., Van Nortrand Reinhold, New York.

Tenijenhuis, K., 1990, The dynamic moduli and the first normal stress difference of the test fluid M1: A combination of results of measurements by various research groups, *J. Non-Newtonian Fluid Mech.* **35**, 169.

Tirumkudulu, M and W.B. Russel, 2003, On the measurement of tack for adhesives, *Phys. Fluids* **15**, 1588.

Tirumkudulu, M., W.B. Russel and T.J. Huang, 2003, Measuring the tack of waterborne adhesives, J. Rheol. 1399

Vlad, D.H. and J.V., Maher, 2000, Tip-splitting instabilities in the channel Saffman-Taylor flow of constant viscosity elastic fluids, *Rhys. Rev. E.* **61**, 5439.