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# Comparative theoretical and experimental studies of breakup, outgassing and stress-induced cavitation of Newtonian and polymerically thickened liquids

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We proposed studies of how liquids fragment under stresses induced by gas flows especially under extreme conditions of ultra high Weber numbers and ultra low pressures. The studies fall broadly into two categories: breakup due to flow induced stresses generated by

- Rayleigh-Taylor instability
- Kelvin-Helmholz instability

and breakup associated also with thermodynamic conditions of the liquids in which outgassing of dissolved gases and liquid vapors play a role. The second category of studies will focus on

- Breakup due to stress-induced cavitation in which flow puts the liquid into tension
- Breakup due to outgassing (cavitation of dissolved gases) especially in polymerically thickened liquids
- Generating systematic experimental data to prove that polymer additives increase the solubility of gases in thickened liquids

## ▪ Rayleigh-Taylor and Kelvin-Helmholtz instability

Our proposed studies are rather specialized; they are in ranges of parameters not previously investigated or they focus on topics for which, to our knowledge, there is no prior literature. Three such topics are: (1) application of viscous and viscoelastic potential flow to Kelvin-Helmholz instability, (2) stress induced cavitation and (3) solubility of gases in thickened liquids.

> **D.D. Joseph, J. Belanger and G.S. Beavers, "Breakup of a liquid drop suddenly exposed to a high-speed airstream,"** ***Int. J. Multiphase Flow*****, 25 (6-7), 1263-1303 (1999).**
> The breakup of viscous and viscoelastic drops in the high speed airstream behind a shock wave in a shock tube was photographed with a rotating drum camera giving one photograph every 5μs. From these photographs we created movies of the fragmentation history of viscous drops of widely varying viscosity, and viscoelastic drops, at very high Weber and Reynolds numbers. Drops of the order of one millimeter are reduced to droplet clouds and possibly to vapor in times less than 500μs. The movies … reveal sequences of breakup events which were previously unavailable for study. Bag and bag-and-stamen breakup can be seen at very high Weber numbers, in the regime of breakup previously called "catastrophic." The movies allow us to generate precise displacement-time graphs from which accurate values of acceleration (of orders $10^4$ to $10^5$ times the acceleration of gravity) are computed. These large accelerations from gas to liquid put the flattened drops at high risk to Rayleigh-Taylor instabilities. The most unstable Rayleigh-Taylor wave fits nearly perfectly with waves measured on enhanced images of drops from the movies, but the effects of viscosity cannot be neglected. Other features of drop breakup under extreme conditions, not treated here, are available on our Web site.

The application of Rayleigh-Taylor instability analysis to our experiments on the breakup of viscoelastic drops in a high-speed airstream is described in the following paper.

> **D.D. Joseph, G.S. Beavers and T. Funada, "Rayleigh-Taylor instability of viscoelastic drops at high Weber numbers," accepted for publication,** ***J. Fluid Mechanics*****.**
> Movies of the breakup of viscous and viscoelastic drops in the high speed airstream behind a

shock wave in a shock tube have been reported by Joseph, Belanger and Beavers [1999]. A Rayleigh-Taylor stability analysis for the initial breakup of a drop of Newtonian liquid was presented in that paper. The movies … show that for the conditions under which the experiments were carried out the drops were subjected to initial accelerations of orders $10^4$ to $10^5$ times the acceleration of gravity. In the Newtonian analysis of Joseph, Belanger and Beavers the most unstable Rayleigh-Taylor wave fits nearly perfectly with waves measured on enhanced images of drops from the movies, but the effects of viscosity cannot be neglected. Here we construct a Rayleigh-Taylor stability analysis for an Oldroyd B fluid using measured data for acceleration, density, viscosity and relaxation time $\lambda_1$. The most unstable wave is a sensitive function of the retardation time $\lambda_2$ which fits experiments when $\lambda_2/\lambda_1 = O(10^{-3})$. The growth rates for the most unstable wave are much larger than for the comparable viscous drop which agrees with the surprising fact that the breakup times for viscoelastic drops are shorter. We construct an approximate analysis of Rayleigh-Taylor instability based on viscoelastic potential flow which gives rise to nearly the same dispersion relation as the unapproximated analysis.

Our work to date on Kelvin-Helmholtz instability is described in the following paper:

**T. Funada and D.D. Joseph "Viscous potential flow analysis of Kelvin-Helmholtz instability in a channel," accepted for publication, *J. Fluid Mechanics*.**
We study the stability of stratified gas-liquid flow in a horizontal rectangular channel using viscous potential flow. The analysis leads to an explicit dispersion relation in which the effects of surface tension and viscosity on the normal stress are not neglected but the effect of shear stresses are neglected. Formulas for the growth rates, wave speeds and neutral stability curve are given in general and applied to experiments in air-water flows. The effects of surface tension are always important and actually determine the stability limits for the cases in which the volume fraction of gas is not too small. The stability criterion for viscous potential flow is expressed by a critical value of the relative velocity. The maximum critical value is when the viscosity ratio is equal to the density ratio; surprisingly the neutral curve for this viscous fluid is the same as the neutral curve for inviscid fluids. The maximum critical value of the velocity overall viscous fluids is given by inviscid fluids. For air at 20°C and liquids with density $\rho = 1$ g/cc the liquid viscosity for the critical conditions is 15 cp; the critical velocity for liquids with viscosities larger than 15 cp are only slightly smaller but the critical velocity for liquids with viscosities smaller than 15 cp, like water, can be much lower. The viscosity of the liquid has a strong affect on the growth rate. The viscous potential flow theory fits the experimental data for air and water well when the gas fraction is greater than about 70%.

To apply the work on Kelvin-Helmholtz instability to the breakup of thickened liquid we need to do the analysis for viscoelastic liquid.

We (Joseph with Clara Mata and Eduardo Pereyra of PDVSA) applied the analysis of Funada and Joseph (above) to the problem of stability of stratified gas-liquid flow and compared their results with those of other leaders in this field. The results are excellent as shown in Figure 1 below. These results are now being prepared for publication.

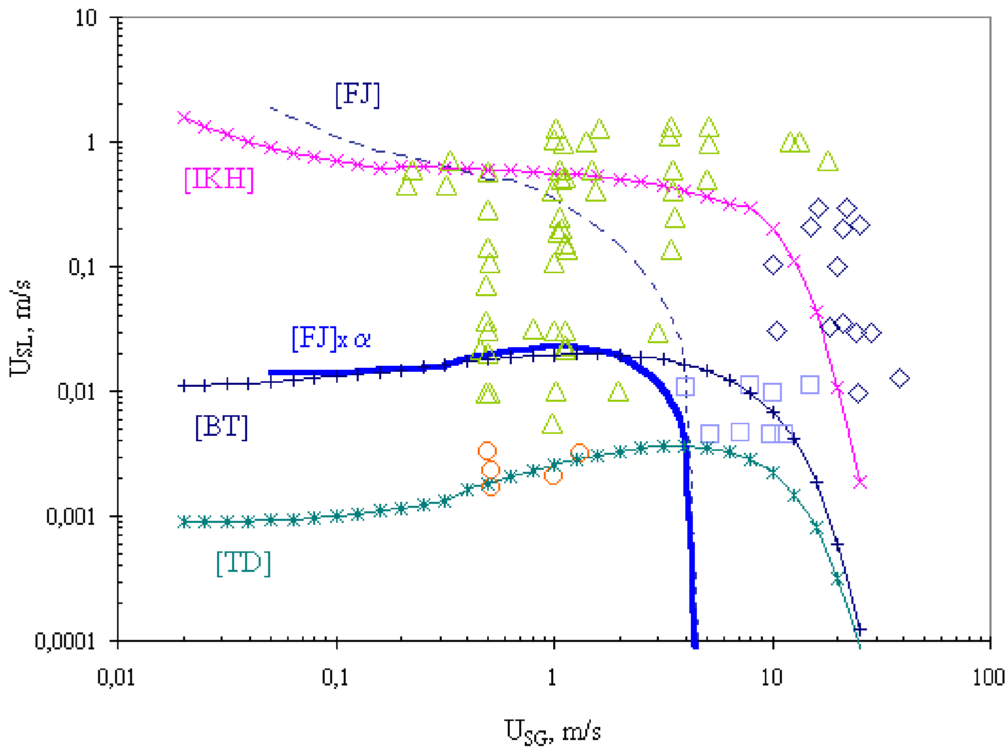



*Figure 1. Neutral curves and experimental data for Kelvin-Helmholz instability (from Stability of stratified gas-liquid flows by Pereyra, Mata and D. D. Joseph, under preparation). Manhane flow chart for PDVSA-Intevep data from 2″ flow loop with air and 480 cP mineral oil. The identified flow patterns are smooth stratified (○ open circles), wavy stratified (□ open squares), slug (Δ open triangles) and annular (◊ open diamonds). Stratified- and non-stratified transition theories after different authors are compared; Taitel and Dukler [TD] 1976 (* stars), Barnea and Taitel [BT] 1993 (+), inviscid Kelvin-Helmholtz [IKH] with $\hat{\mu} = \hat{\rho}$ (×), Funada and Joseph [FJ] 2001 (- - - broken line), Funada and Joseph multiplied by α [FJ]xα 2001(▬ heavy line).*

## Stress induced cavitation

Progress was made on stress induced cavitation in journal bearings, on stress induced rupture of liquid filaments and on cavitation in pure shear.

- ### Cavitation in journal bearings

**A. Pereyra, G. McGrath and D.D. Joseph. Flow and stress induced cavitation in a journal bearing with axial throughput. Accepted for publication, *J. Tribology.***
The problem of predicting flow between rotating eccentric cylinders with axial throughput is studied. The system models a device used to test the stability of emulsions against changes in drop size distribution. The analysis looks for the major variation in flow properties which could put an emulsion at risk due to coalescence or breakage and finds the most likely candidate in the pressure gradient defined as the ratio of the difference between the maximum and minimum pressure to the arc length between the difference. The axial throughput is modeled by flow driven by a constant pressure gradient. The flow is calculated from the Navier-Stokes equation using the code SIMPLER (Patankar 1980). The effects of inertia at values typical for the device are studied. Several eccentricities and different rotational speeds are computed to sample the changes in flow and stress parameters in the idealized device for typical conditions. The numerical analysis is

validated against the lubrication approximation in the low Reynolds number case. Conditions for stress induced cavitation are evaluated.

The flow is completely determined by a Reynolds number, an eccentricity ratio and a dimensionless pressure gradient and all computed results are either presented or can be easily expressed in terms of these dimensionless parameters.

The effect of inertia is to shift the eddy or re-circulation zone which develops in the more open region of the gap toward the region of low relative pressure; the zero of the relative pressure migrates away from the center and the distribution breaks the skew symmetry of the Stokes flow solution.

The state of stress in the journal bearing is analyzed and a cavitation criterion based on the maximum tensile stress is compared with the traditional criterion based on pressure.

- **Stress induced rupture of liquid filaments**

Lundgren and Joseph did a heuristic analysis of the breakup of a liquid capillary filament using viscous potential near a stagnation point on the centerline of the filament towards which the surface collapses under the action of surface tension forces. They found that the neck is of parabolic shape and its radius collapsed to zero in a finite time. During the collapse the tensile stress due to viscosity increases in value until at a certain finite radius, which is about 1.5 microns for water in air, the stress in the throat passes into tension, presumably inducing a cavitation there.

The main points about the analysis of Lundgren and Joseph are that in the collapse in finite time, the extensional viscous stress goes to infinity, and the pressure drops due to the high velocity at collapse, a Bernoulli effect. Both effects favor cavitation as the mode of collapse. We want to prove this.

The main feature leading to capillary rupture due to cavitation is the collapse of the radius in a finite time which is linear in $(t\text{-}t_0)^{-1}$. This feature comes also out of different types of analysis by Eggers (1993) and Papageorgiou (1995), and it is in agreement with McKinley and Tripathi (2000) and Anna and McKinley (2001). The only difference among these authors is in the prefactor of the linear collapse law.

Our goal is to show conclusively that all the theories lead to rupture by cavitation. Funada and Joseph (2001) have finished but not published an analysis of the linear theory of capillary instability. They showed that the results of analysis by viscous potential flow is the same as the fully viscous flow without approximation when a Reynolds number $\frac{\gamma d\rho}{\nu^2}$, where $\gamma$ is surface tension, $d$ is the undisturbed cylinder diameter, $\rho$ and $\nu$ are the density and kinematic viscosity of the liquid (collapsing in air), is large enough. This criterion includes most practical cases and it reinforces the idea that viscous potential flow is the right frame for a nonlinear analysis of capillary breakup and rupture. We plan such an analysis using numerical methods.

- **Cavitation in pure shear**

Joseph (1998) predicted that cavities would open in pure shear when the largest principal stress exceeded a cavitation. An experiment realizing this prediction was reported in a paper by Archer, Ternet, and Larson 1997: “Fracture” phenomena in shearing flow of viscous liquids. They note that

> …the shear stress catastrophically collapses if the shear rate is raised above a value corresponding to a critical initial shear stress of around 0.1-0.3 Mpa. …in polystyrene, bubbles open up within the sample; as occurs in cavitation. Some similarities are pointed out between these phenomena and that of ‘lubrication failure’ reported in the tribology literature.

The critical stress 0.1-0.3 Mpa = 1-3 atmospheres is just what might have been guessed for cavitation under shear.

Even earlier Winer and Bair (1987) and Bair and Winer (1992) did experiments with high viscosity liquids between concentric cylinders and they looked at rheology at high stress. Winar and Bair (1987) concluded that

> … The tentative conclusion of this work is that the pseudoplastic behavior of some liquids is apparently the result of the reduction of the principal normal stress at high shear stress causing void formation and the reduction of apparent viscosity.
>
> However, for some high shear rate viscosity data at atmospheric pressure the principal normal stress may approach quite low values relative to 1 atmosphere suggesting the possibility of cavitation or fracture of the material resulting in a reduced shear stress.
>
> … Because this type of stress field exists in elastohydrodynamic inlets and classical hydrodynamic configuration, such as high speed journal bearings, it should be further investigated, because it potentially represents a lubricant limitation to feeding bearings. It also may be an influence on the location of the cavitation boundary at the exit of hydrodynamic films. This mechanism should be explored further as a possible cavitation boundary condition to hydrodynamic lubrication.

Taken together, these experiments do establish that cavitation is associated with the principal state of stress rather than the pressure (the mean normal stress).

## Solubility and Outgassing

We first encountered the problem of the outgassing of certain liquids in which a polymer had been dissolved when we tried to carry out aerodynamic breakup studies at low ambient pressure in our shock tube. We were studying the breakup of tributyl phosphate (TBP) that had been thickened with polystyrene butyl acrylate (PSBA) in various concentrations. The TBP/PSBA solutions outgassed violently into a very unstable foam at pressures near 1 torr, a pressure that is much higher than the vapor pressure of neat TBP (0.014 torr). Based on our early observations we proposed the following wide-ranging program:

***Solubility of thickened liquids.*** *Find the weight fraction of dissolved air in thickened liquids as a function of temperature, pressure, polymer concentration, and polymer molecular weight.*

We are carrying out an extensive experimental program along these lines, presently working exclusively with TBP because of its very low vapor pressure. We are using a variety of polymers listed in the table below:

| | Molecular Weight (MW) | Concentration (%) |
|---|---|---|
| PSBA | $3.0 \times 10^6$ | several between 1 and 9 |
| PSBMA 90% Styrene | $1.30 \times 10^6$ | 3.0, 4.8 |
| PSBMA 90% Styrene | $2.87 \times 10^6$ | 3.0, 4.8 |
| PSBMA 10% Styrene | $2.27 \times 10^6$ | 3.0, 4.8 |
| PVB | 320,000 | 3.0 |
| PBMA | 337,000 | 3.0, 4.8 |
| PIBMA | $3.77 \times 10^6$ | 3.0, 4.8 |

*Table 1.*

Gas solubility increases with polymer concentration, as illustrated in Figure 2 for TBP/PSBA. We are still trying to isolate the effects of molecular weight and polymer composition.

We are completing experiments that appear to demonstrate that the absorbed gas is in fact water vapor. Extensive experiments with neat TBP on the outgassing and re-absorption in dry and humid environments are all consistent with water vapor as the absorbed gas. Similar experiments are continuing with thickened TBP. The next phase is to examine solubility effects in another low-vapor-pressure liquid, namely dibutyl phosphate. This work is on-going and will be written up for publication when all the results are definitive.

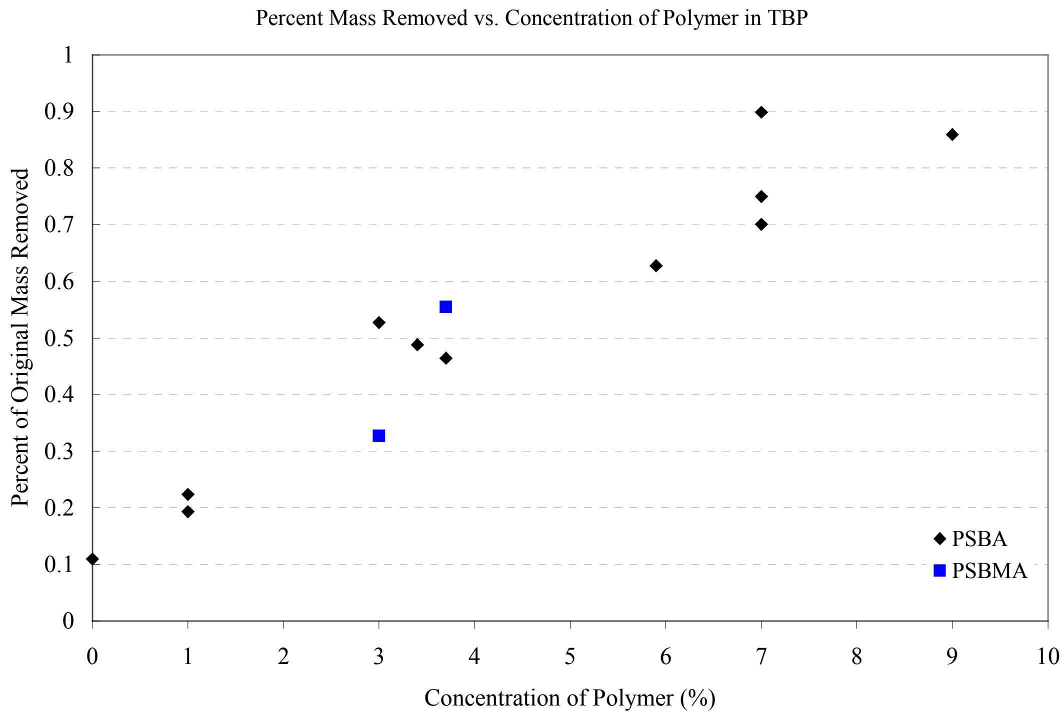
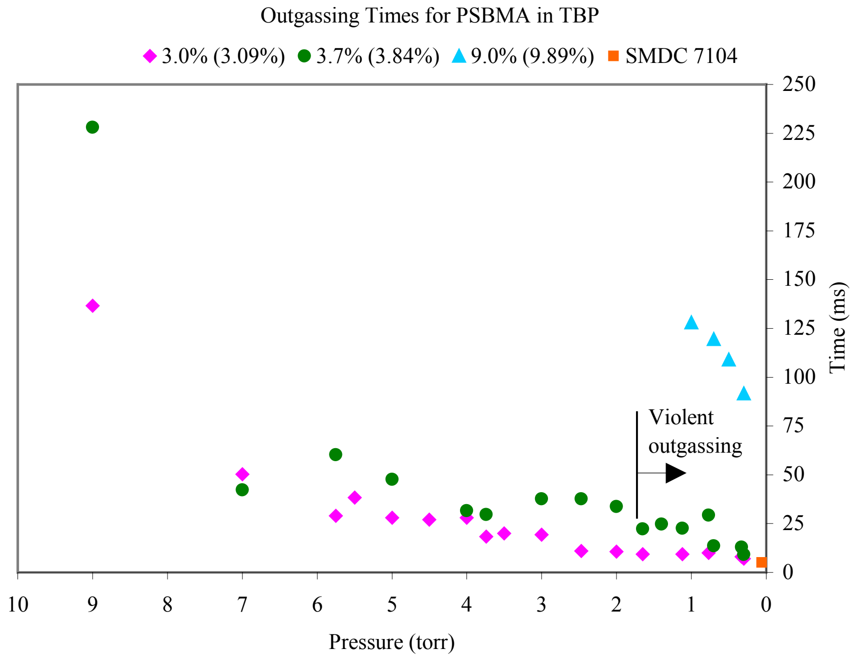
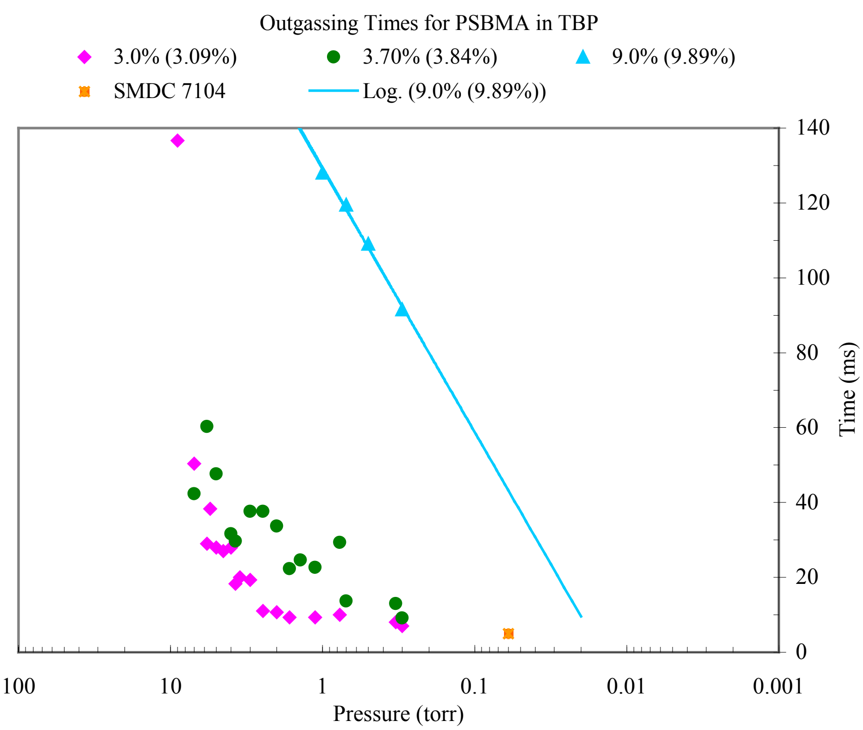



*Figure 2. Percent mass removed vs. concentration of polymer in TBP.*




(a)




(b)

*Figure 3. Time to start outgassing vs. pressure for different concentrations of TBP/PSBMA (90% styrene, 1.3 million Mol. Wt.).*

In parallel with the solubility experiments we are measuring the time interval between exposure to a low-pressure environment and the instant at which explosive outgassing begins. This depends strongly upon viscosity, which in turn depends on the polymer type and concentration. Typical times to start outgassing are shown in Figure 3 for several concentrations of PSBMA in TBP. These data are important for aerodynamic breakup experiments in low ambient pressures.

## Aerodynamic Breakup

We have recently captured on film Rayleigh-Taylor waves on a droplet of an aqueous solution of polyox in the flow behind a shock Mach number of 3.0 corresponding to a free stream dynamic pressure of approximately 600 kPa. These waves are most distinct and most easily identifiable in a small interval (about 15 microseconds) that starts about 30 microseconds after the drop is exposed to the high speed flow and during which the drop is undergoing a severe flattening of both the front and rear faces. Figure 4 shows contrast-enhanced pictures at 5 microsecond intervals. The wavelengths are readily measured from the computer screen at 5 pixels per wave on the scale at which the pictures were analyzed.

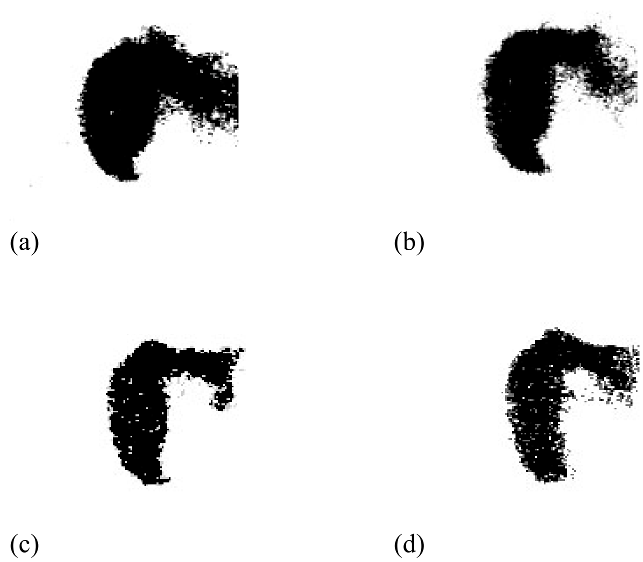
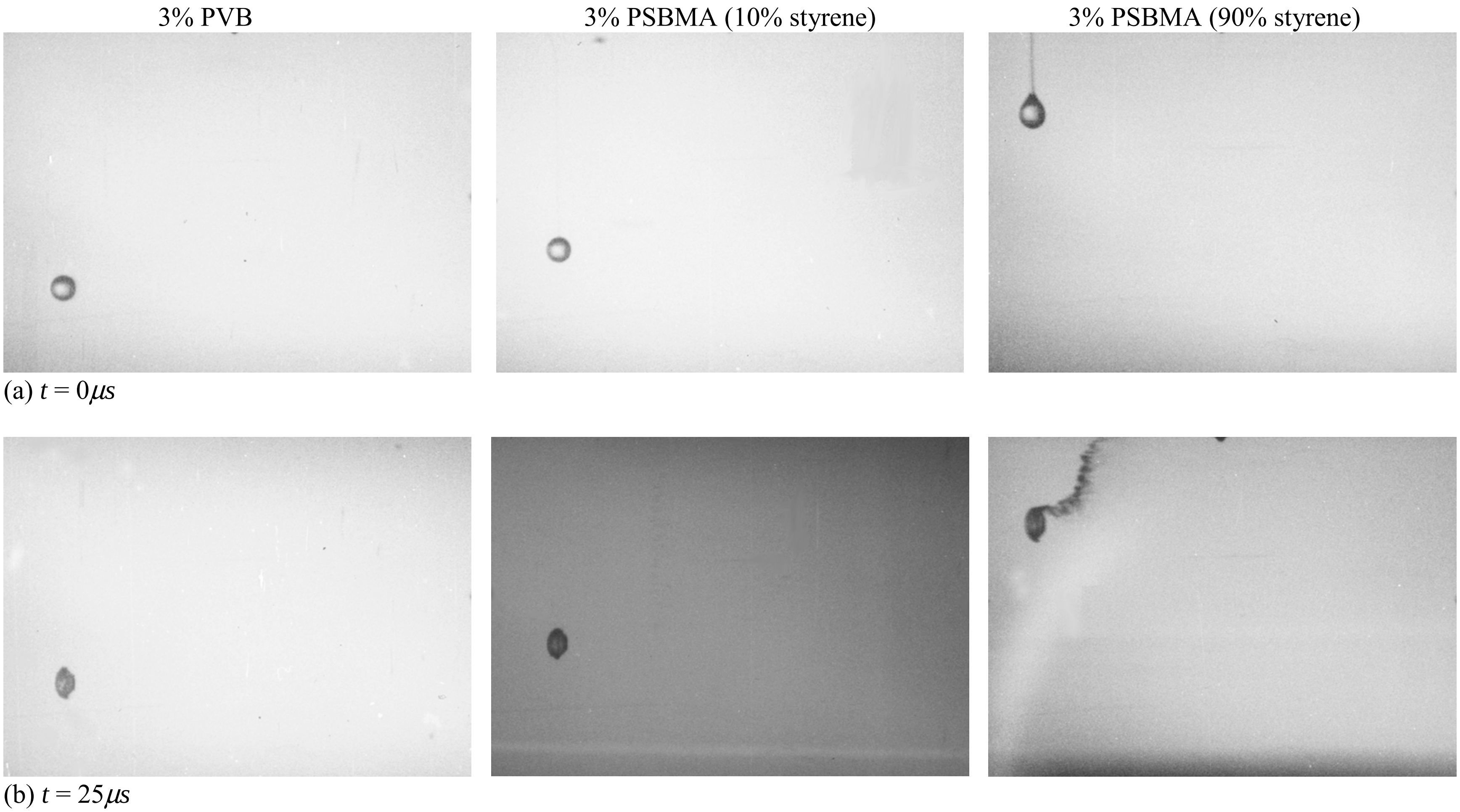


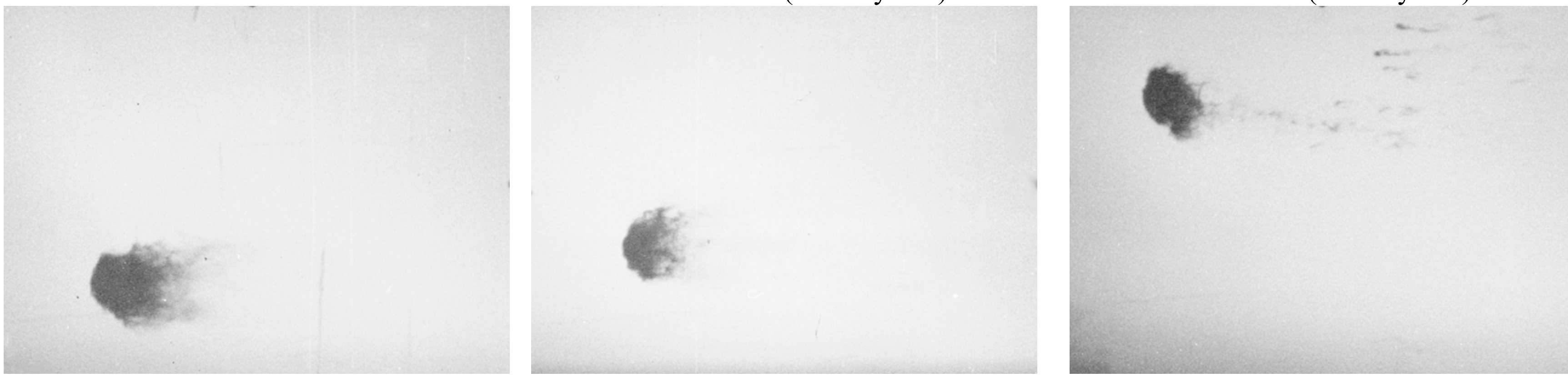

*Figure 4. Contrast-enhanced images of a drop of 2% aqueous polyox undergoing deformation, showing the Rayleigh-Taylor waves on the front surface. Time from initial exposure to flow in microseconds: (a) 30 (b) 35 (c) 40 (d) 45.*

Breakup studies are currently being carried at a fixed shock Mach number of 3.5 and two different free stream dynamic pressures, namely 660 kPa and 220 kPa, to determine the influence of polymer type and concentration on breakup characteristics. Figure 5 shows a typical comparison for TBP thickened with 3.0% of different polymers. This figure is constructed from shots to determine the effect, if any, that styrene content in the thickener might have on aerodynamic breakup times and resultant debris field. These experiments are still on-going. As a complement to these studies we are attempting breakup shots under the above conditions using four different concentrations of PBMA (Mol. Wt of about 2.6 million) in TBP. This aerodynamic breakup program is far from complete and it will probably be another year or so before sufficient definitive data are ready for publication.

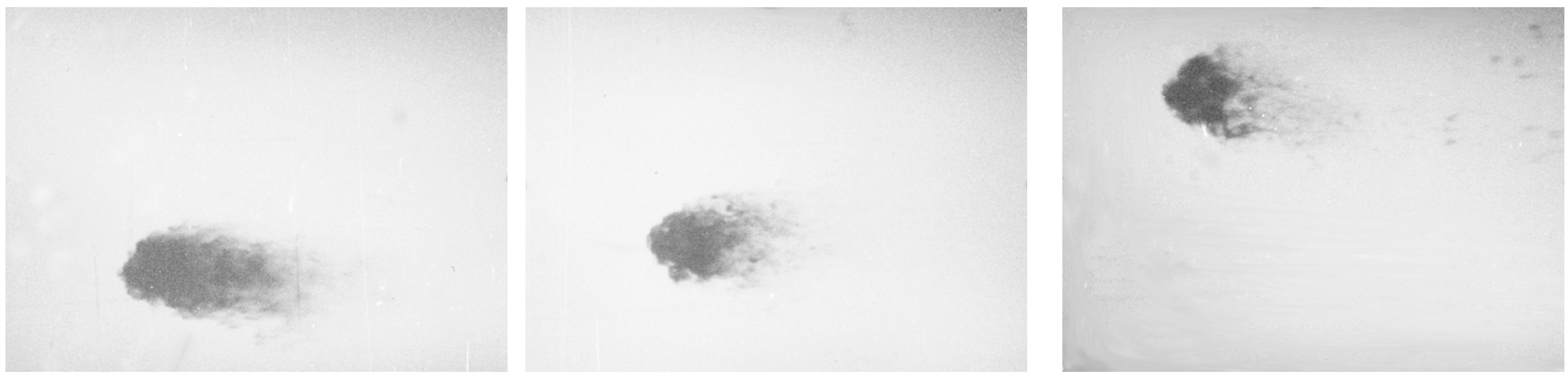
*Figure 5(a-f). Comparison of the aerodynamic breakup of three liquids with varying styrene content at the same conditions (q = 660 kPa, Mx = 3.5).*



3% PVB | 3% PSBMA (10% styrene) | 3% PSBMA (90% styrene)

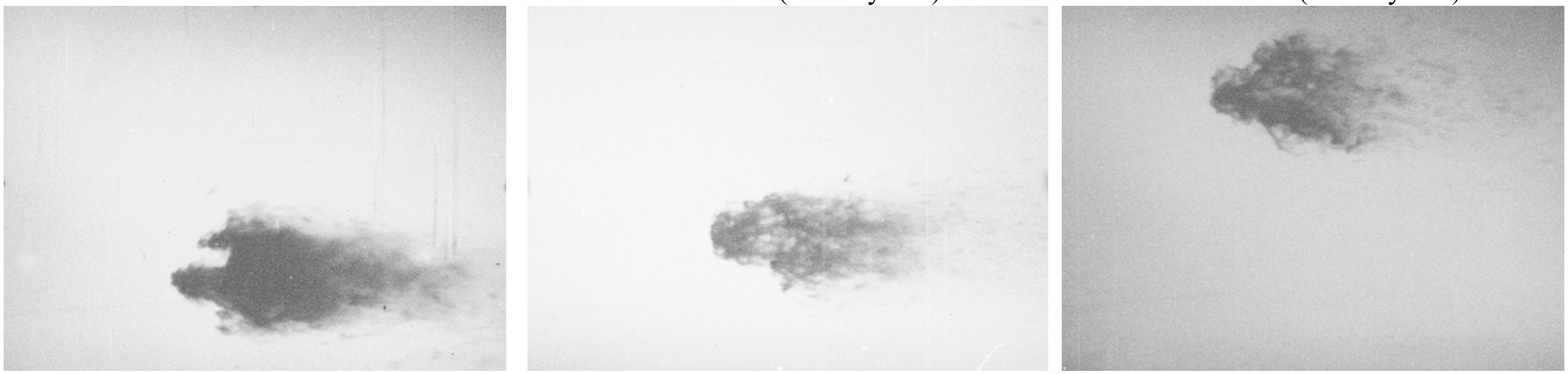


(c) $t = 75\mu s$

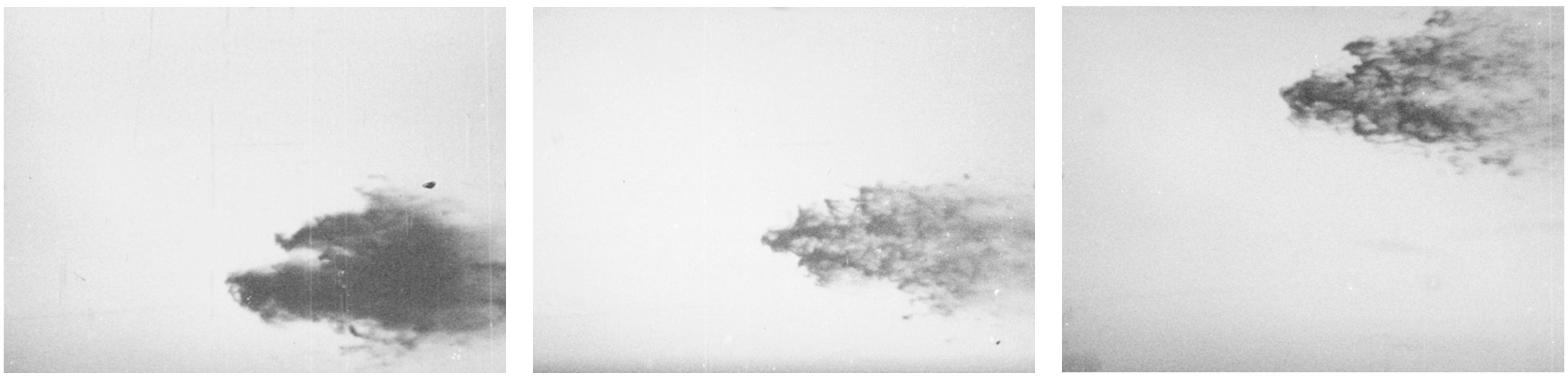


(d) $t = 100\mu s$

3% PVB | 3% PSBMA (10% styrene) | 3% PSBMA (90% styrene)



(e) $t = 130\mu s$



(f) $t = 155\mu s$

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