

RHEOLOGICAL STUDIES OF EMULSION INK STABILITY

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ABSTRACT

Rheological measurements of emulsion inks were done using a controlled-stress rheometer with a cone and plate geometry and a controlled-rate rheometer with a concentric cylinder geometry. The results from both experiments were consistent. The emulsion inks can be classified into stable, metastable, and unstable emulsions according to the shape of flow curves and viscosity versus temperature curve.

Flow curves of stable emulsion inks are very similar to those of fresh inks without added fountain solution. These curves are typical of pseudo-plastic systems. They feature a yield value at zero shear rate and a gradual decrease in the viscosity with increasing shear rate.

Flow curves of metastable emulsions become irregular at higher shear rates where the viscosity changes randomly with increasing shear rate. These observations are consistent with the assumption of incipient water separation and a concurrent reformation of emulsion.

A shear-induced gross water separation characterizes unstable emulsion inks. Slippage between the moving surfaces of measuring systems taking place in the newly formed water layer causes an apparent loss of viscosity. This results in the shear rate jumping to infinity at a certain shear stress with the controlled-stress rheometer. Correspondingly, the shear stress drops to nearly zero as the applied shear rate exceeds a certain value with the controlled-rate rheometer.

The results indicate that the stability of emulsion inks is dependent upon the physico-chemical nature of inks and fountain solutions, the composition of emulsion inks, and the temperature. A given system may behave as a metastable or a stable emulsion depending upon the shear history of the sample. These factors will be discussed in this paper.

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BACKGROUND

The lithographic printing process differs from other printing processes in that both image and non-image areas are on the same plane. So, image differentiation requires in addition to the ink an aqueous fountain solution. These two immiscible fluids are separately applied to the printing plate through different distribution systems. After application free water may be present on top of the image areas and on the surface of ink form rollers. It can interfere with subsequent ink transfer. To ensure proper ink transfer, lithographic inks are so designed that they can absorb excess fountain solution to form an emulsion. The water content of ink on form rollers can range from 3% for a metal decorating ink to 47% for a newspaper run on a sheet-fed press (Fetsko, 1986). Therefore, lithographic presses print a water-in-ink emulsion rather than a dry ink. The time for the emulsification process to reach an equilibrium state is probably the major cause of the printing waste.

Many researchers studied emulsification properties of inks in the laboratory to predict their performance on the press (Surland, 1980 and 1983, and Tasker et al., 1983). Surland used a modified Sunbeam Mixmaster to emulsify fountain solution into an ink in a time frame of ten minutes. The quantity of emulsified fountain solution was measured every minute to establish an emulsification curve. Press performance was predicted on the basis of the shape of the emulsification curve. Though this method had succeeded in many instances, Konięcki and co-workers (1985) pointed out that it might not be reliable. Fountain solution being emulsified could vary with the quantity of fountain solution added before mixing (Chou et al., 1987).

Tasker and coworkers (1983) conducted emulsification tests on a Litho-Break Tester that better simulates real press conditions. The quantity of emulsified fountain solution was measured as a function of time. They treated emulsification as a pseudo-first-order reaction. This model allows them to calculate the apparent emulsification rate constant and the ultimate water-pickup. Their experiments showed that some ink/fountain solution pairs had similar emulsification properties but differed significantly in press performance.

Emulsification tests provide a means of determining the emulsification rate and the emulsification capacity of an ink.

They do not measure changes in physical properties of the ink caused by emulsified fountain solution. This is probably one of the reasons why these tests are not enough to predict ink performance.

Rheology is the most important physical property of inks that determines press performance. However, the effect of emulsification on rheological properties of inks is not well understood. Tasker and co-workers (1983) used a Laray viscometer to measure the change in plastic viscosity, the viscosity at shear rate of 2500 sec^{-1} . They noted that a large change in plastic viscosity due to emulsification could cause printing problems. Bassemir and Shubert (1985) used similar techniques to determine the plastic viscosity and the yield value as a function of water content. Yield value is the shear stress extrapolated to a shear rate of 2.5 sec^{-1} . The results were reported as shortness ratio, the ratio of yield value to plastic viscosity. They concluded that inks would perform poorly on press if emulsification resulted in an excessively large increase in shortness ratio.

In the study of emulsion ink rheology using a more sophisticated instrument, Chou and Fadner (1986) observed that emulsion inks containing 20% fountain solution or more broke down at shear rates much lower than 2500 sec^{-1} . It is therefore impossible to determine the "plastic viscosities" of those emulsion inks. And, the determination of changes in plastic viscosity and shortness ratio caused by emulsification is also impossible. However, their results showed the dependence of emulsion ink stability upon the physico-chemical nature of inks and fountain solutions. The stability of emulsion inks increased with decreasing water content and with increasing temperature. Emulsion inks that were too stable or too unstable did not perform satisfactorily on their keyless test press.

Chou and Bain (1988) also made extensive studies on the rheology of fresh newsinks. Those inks behaved more or less alike at high shear rates due to diminishing internal structure. However, they differed significantly in rheology at shear rates less than a few hundred reciprocal seconds. The results show that keyless inks have lower dynamic yield values and smaller shear thinning indices than conventional inks of the same consistency. Consequently, they re-investigated the rheological properties of emulsion inks in this shear rate range. Experimental results reported here demonstrate that

viscometry is a very sensitive technique for studying the stability of emulsion inks. No attempt is made to analyze quantitatively the experimental results at the present stage. A systematic and quantitative study is in progress and the results will be reported later.

EXPERIMENTAL

Materials

Four black newsinks and two fountain solutions were used in this study. Ink A obtained from a Japanese source is a conventional offset ink. Inks B, C, and D supplied by two U.S. ink manufacturers are designed for our keyless press. FS1, an alkaline fountain solution, and FS2, a neutral solution, were obtained from two different suppliers. Fountain solutions were diluted to the concentrations specified by the suppliers.

Inks without added fountain solution are referred to in this paper as fresh inks, and inks with added fountain solution as emulsion inks. Emulsion inks were prepared with a high speed mixer by gradually adding a predetermined quantity of fountain solution to the ink.

Rheometry

Two rheometers were used in this experiment. One is a Carri-Med controlled-stress rheometer with a cone and plate geometry. A 4-centimeter, 1.5-degree cone was used for studying flow behavior at shear stresses up to 5900 dynes/cm². A 2-centimeter, 1.5-degree cone was used for higher shear stresses. The other is a controlled-rate Haake RV20 Rotovisco rheometer with a concentric cylinder geometry. The diameters of bob and cup are 4.008 and 4.2 centimeters, respectively. The maximum allowable shear stress in the controlled-rate rheometry is 3200 dynes/cm².

Two techniques were used to investigate the rheological behavior of inks. In flow measurements, either the shear stress or shear rate, depending on the instrument, increased linearly from zero to a programmed maximum value and then decreased in the same way back to zero. The resultant shear rates or shear stresses were measured. This method results in up and down flow curves. All flow measurements were done at 25° C, except otherwise mentioned.

In temperature sweep measurements, a constant shear stress or shear rate was applied to the sample, while temperature was gradually decreased from about 45° C to a lower value. The shear rates or shear stresses were recorded as a function of temperature.

Because the rheological properties of a fluid depend strongly on its shear history, ink samples were presheared at five reciprocal seconds for one minute. They were then left undisturbed for five minutes before the start of measurement. We observed that this method gave reproducible results and hardly affected the internal structure of the sample.

RESULTS AND DISCUSSIONS

Flow Curves of Fresh Inks

The shear stress versus shear rate flow curves shown in Figure 1 are very similar for both rheometer measuring systems. These curves are typical of fresh inks and feature relatively smooth up and down curves. Their slopes decrease gradually with increasing shear rate. Inks may or may not exhibit yield values at zero shear rate.

The decreasing slope of the up curve with increasing shear rate corresponds to a decreasing viscosity. This is due to the internal structure of the ink being progressively destroyed by the increasing shear force. Its resistance to flow decreases correspondingly. As the shear rate decreases, the internal structure starts to reform but is unable to completely recover. So, the down curve appears below the up curve and its slope increases with decreasing shear rate. This results in the well-known hysteresis loop. The curvature of the down curve is smaller than that of the up curve for all fresh inks. This behavior also suggests an incomplete recovery of internal structure.

Curvature details of these flow curves characterize the shear-dependent (shear thinning) property of the fluid or its deviation from Newtonian behavior. The area enclosed in the hysteresis loop is a measure of a time-dependent property of the fluid known as thixotropy. The viscosity of a thixotropic fluid decreases with time under a steady shear rate. Both properties are known to be related to the internal structure of inks (Chou and Bain, 1988).

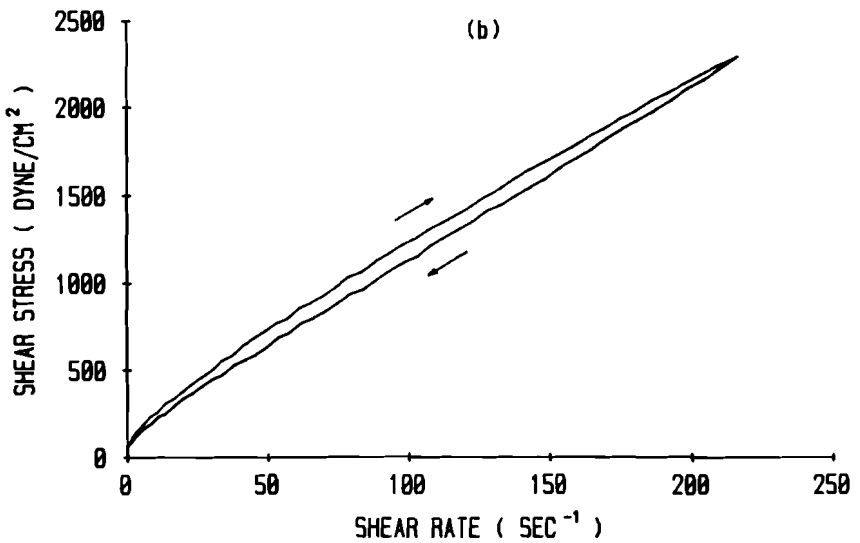
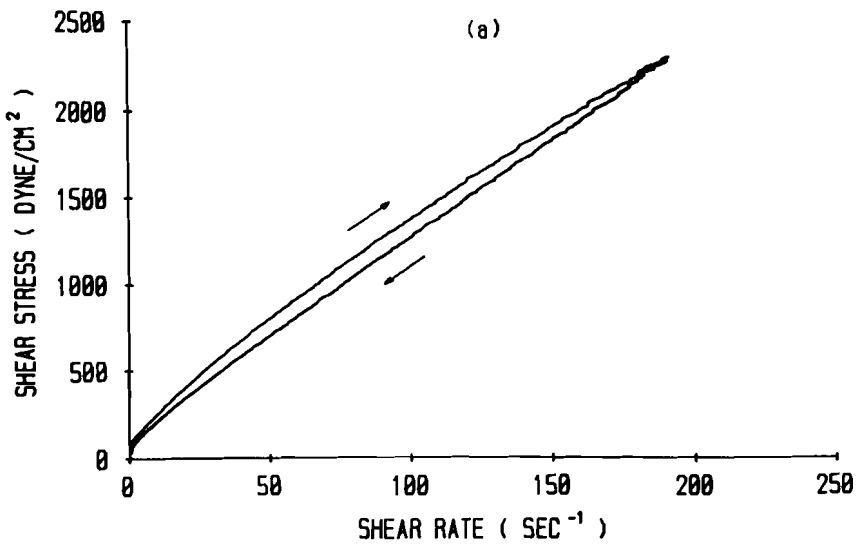


Figure 1. Flow curves of fresh ink B from (a) the controlled-stress rheometry and (b) the controlled-rate rheometry.

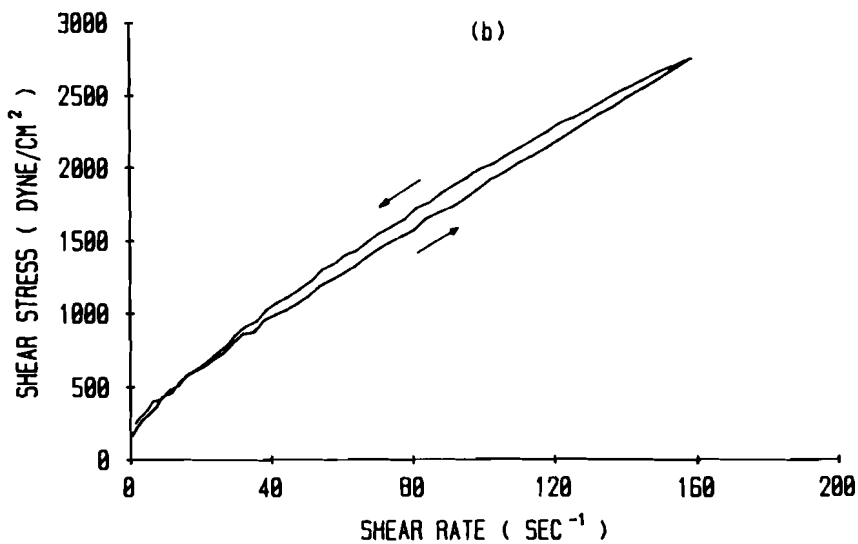
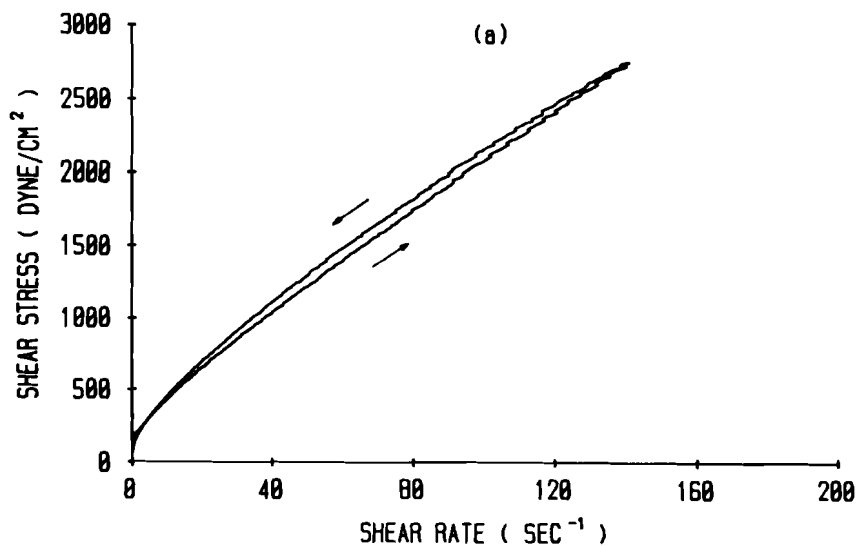


Figure 2. Flow curves of emulsion ink B containing 40% FS1 from (a) the controlled-stress rheometry and (b) the controlled-rate rheometry.

Flow Curves of Emulsion Inks

Figure 2 shows the flow curves of ink B containing 40% fountain solution FS1. These curves are again very similar for the two instruments. Both the up and down curves are as smooth as those of fresh inks, typifying stable emulsion inks. It is unusual that the up curve falls below the down curve. This type of rheological behavior is referred to as rheopexy (Patton, 1979), opposite of thixotropy. Possible reasons for rheoplectic behavior in emulsion inks will be discussed later.

Figure 3 shows an example of flow curves for a metastable emulsion ink. The up curve is initially as smooth as that of fresh inks but become irregular at higher shear rates. The irregularity continues during the downward measurement but eventually disappears. The down curve regains a smooth shape.

Figure 4 illustrates flow curves typical of unstable emulsion inks. These curves have characteristics similar to metastable emulsions at low shear rates. However, the shear rate suddenly jumps to infinity at a certain shear stress when using the controlled-stress rheometer. Correspondingly, the shear stress drops to nearly zero as the applied shear rate exceeds a certain value when using the controlled-rate rheometer.

Stability of emulsions has been reviewed in detail by Rosen (1978) and is briefly discussed here. The dispersion of one liquid into another liquid, for instance water into oil, by a mechanical method produces a tremendously large increase in the interfacial area. The dispersed particles will soon coalesce to reform as a separate phase if the two pure fluids are immiscible. This is because the interfacial tension between them is always greater than zero. However, in the presence of surfactants the dispersion process produces more or less stable emulsions. Each surfactant molecule consists of a hydrophilic and a lyophilic groups. These surfactant molecules adsorb at the interface and orient themselves in a way that the hydrophilic heads point toward the water phase and the lyophilic tails toward the oil phase. This arrangement of molecules forms an interfacial region between the two liquids. It reduces the interfacial free energy of the system and produces barriers around the dispersed particles so that the coalescence rate is significantly reduced and the emulsion appears stable under these conditions.

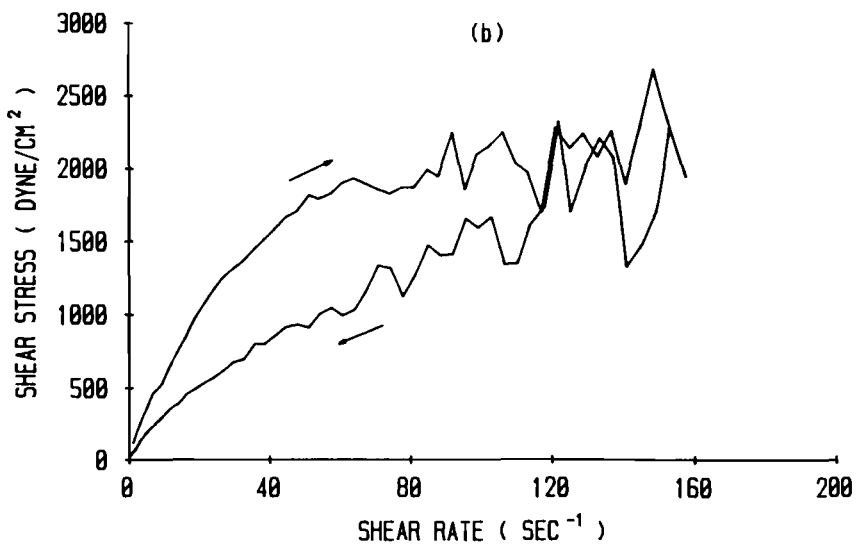
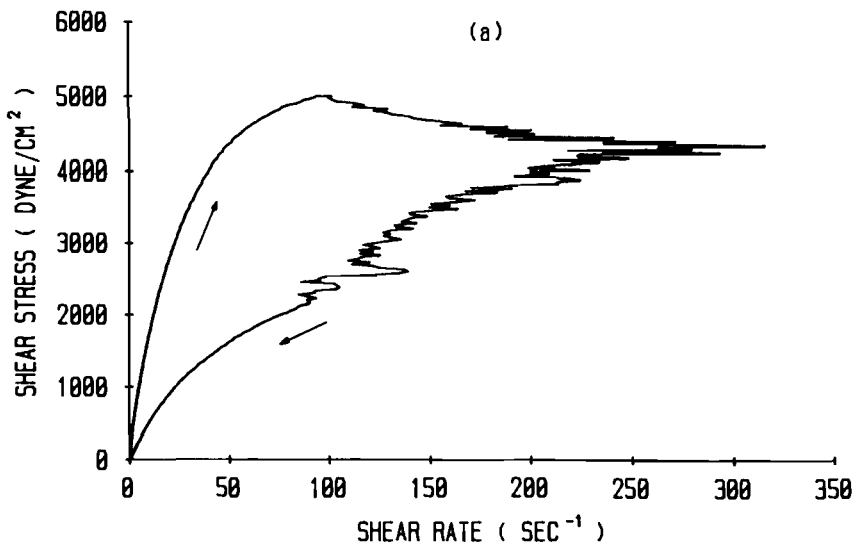


Figure 3. Flow curves of emulsion ink A containing 30% FS1 from (a) the controlled-stress rheometry and (b) the controlled-rate rheometry.

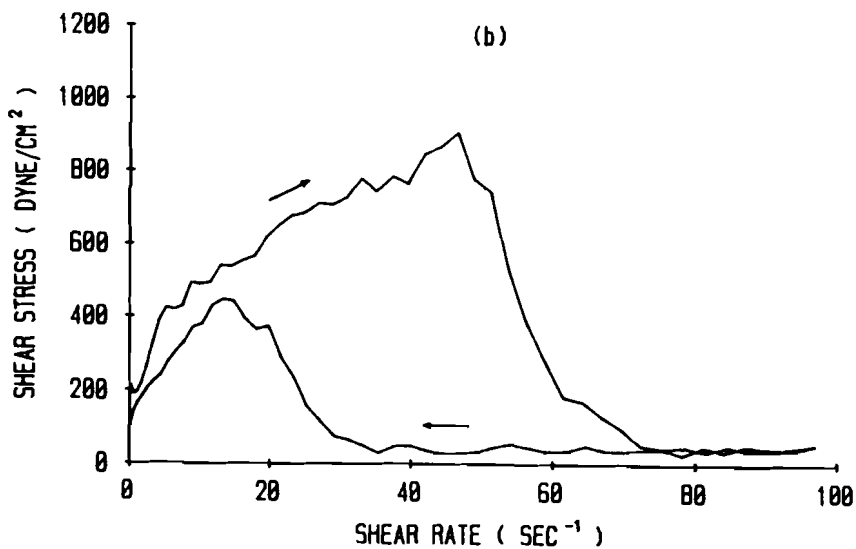
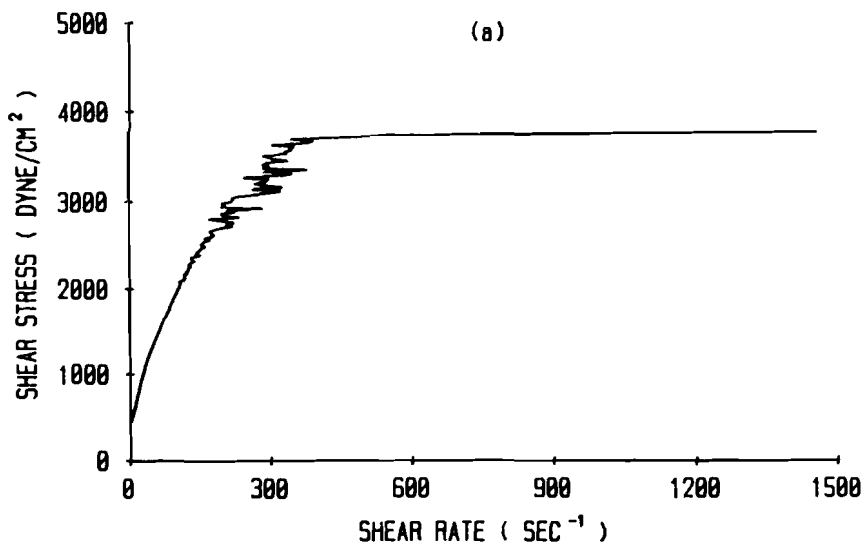


Figure 4. Flow curves of emulsion ink C containing 20% FS1 from (a) the controlled-stress rheometry and (b) the controlled-rate rheometry.

The interfacial tension between the hydrophilic heads and water molecules can be different from that between the lyophilic tails and oil molecules. This difference in the interfacial tensions produces a driving force (interfacial force) that tends to curve the interfacial region in a way that minimizes the area of the interface with greater interfacial tension and in this way reduces the interfacial free energy as required by the laws of thermodynamics. So, the dispersed particles are present in the form of spheres within the continuous phase. Surfactants that are more soluble in the oil than in the water will produce w/o emulsions, and vice versa.

Experimental results from dielectric constant measurements of emulsion inks indicate that emulsified fountain solution is present within the ink matrix in the form of individual spherical droplets under static conditions (Cher et al., 1988). When the emulsion ink is forced to flow, these spherical droplets are deformed by the shear force into prolate spheroids along the stream line of flow. The increased surface area caused by the shear force reduces the stabilizing strength of the interfacial region. It also increases the collision radius of dispersed particles. Both factors increase the coalescence rate of emulsified fountain solution. These two counteracting forces govern the shear stability of emulsion inks.

At low shear rates, the stabilizing interfacial force dominates over the destabilizing shear force. The deformation of dispersed droplets is minimal. The interfacial force is also dominant in stable emulsion systems. Under these conditions, emulsion inks behave rheologically like a homogeneous fluid rather than like a two phase system. So, the resulting flow curves are as smooth as those of fresh inks.

As the shear rate or shear stress increases, the shear force becomes comparable in magnitude to the interfacial force. The dispersed droplets become significantly deformed. This deformation help dispersed particles coalesce into larger droplets or to form discrete water layers. Discrete water layers provide a least-resistance path for flow in response to the applied shear. As a result, the measured shear stress suddenly decreases in controlled-rate rheometry and the shear rate increases instantly in controlled-stress rheometry. These discrete water layers may become so grossly deformed by the shear force that they eventually break up into many small droplets. Re-dispersion thus takes place and the associated viscosity increase is readily seen by the presence of a peak in

the flow curve. This model of incipient water separation from emulsions and concurrent emulsion reformation satisfactorily explains the zigzag appearance of the flow curves. These two processes persist in the downward flow measurement until the shear force decreases sufficiently. The interfacial force then takes over and the down curve eventually becomes smooth again.

For unstable emulsion inks at high shear rates, the shear force becomes overwhelmingly dominant. The coalescence process proceeds until a continuous water layer forms. Slippage between moving surfaces of the measuring devices takes place in this newly formed, very fluid water phase. This causes the shear rate jumping to infinity with the controlled-stress rheometer. Correspondingly, the shear stress drops to nearly zero with the controlled-rate rheometer. This gross water separation characterizes unstable emulsion inks. The controlled-stress rheometer is automatically shut down when this happens and the down curve is unobtainable. The rheological measurement can continue with the controlled-rate rheometer. The down curve is initially parallel to and very close to the shear rate axis. This behavior indicates a region where the water layer still exists. As the interfacial force begins to balance the decreasing shear force, the down curve rises and finally resumes virtually the same pattern as a stable emulsion.

In a previous publication, Chou and Fadner (1986) observed that the shear stability of an emulsion ink decreased with increasing water content under high shear conditions. The number of dispersed particles in an emulsion ink is expected to be proportional to the water content. The collision frequency of water particles increases accordingly with increasing water content. The shear-induced increase in the surface area of dispersed water particles decreases the strength of the interfacial region. These two factors make the coalescence process easier. Unpublished results show that the coalescence rate of emulsion inks is proportional to the square of water content. The indication is that the coalescence process is a bimolecular reaction. This is in agreement with the incipient water separation mechanism proposed here.

Figures 2 to 4 show that ink B containing 40% FS1 is the most stable emulsion ink of the three. Ink C with only 20% FS1 is least stable. These results infer that the shear stability of emulsion inks strongly depends on the physico-chemical nature of the ink and presumably of the fountain solution.

Instability occurs at a shear stress in the controlled-rate rheometer lower than that in the controlled-stress rheometer (Figures 3 and 4). This inconsistency can be ascribed to the different measuring systems. Centrifugal force exists in these measuring systems. This may facilitate the preferential diffusion of water particles to the ink/air interface of the cone and plate rheometer. The appearance of some water droplets at this edge during measurement confirms this model. The number of water particles in the instrument's measuring gap is thereby reduced, so is the coalescence rate. In the concentric cylinder geometry, water particles may be centrifugally trapped and accumulated in a layer close to the non-moving cup surface. The increased concentration of water particles accelerates the incipient water separation process taking place in that region.

Effect of Shear Conditions

The flow curves in Figures 2 to 4 show the transition of emulsion inks from a stable through a metastable to an unstable state as a function of shear rate. Nevertheless, the shear rates above which emulsion inks exhibit instability or breakdown appear much lower than those existing in the high speed mixer container during emulsion ink preparation. This discrepancy can be attributed to the different flow patterns in the two systems, laminar versus turbulent. High shear forces present near blades of a high speed mixer help break large water droplets into smaller ones which are then dispersed radially. Possibilities that two stretched droplets will coalesce are considerably lower under turbulent flow. Phase separation is therefore less likely to happen under these conditions. We observed that in many instances even when inks were over-emulsified, they looked like homogeneous fluids right after mixing. Free water appeared after they were allowed to stand for a while or slightly squeezed by a knife.

All emulsion inks in this study exhibited flow characteristics of stable systems under minor shear conditions. They might break down as the shear force became vigorous. Figure 5 presents an example illustrating this effect. The emulsion ink appears stable up to a maximum shear stress of 3500 dynes/cm² (Figure 5a). It begins to exhibit instability at about 4500 dynes/cm² (Figures 5b). Slight increase in the shear stress to 5000 dynes/cm² results in a significant change in the shape of flow curves, as shown in Figure 3a. Emulsion breakdown occurs at a shear stress less than 8000 dynes/cm² (Figure 5c).

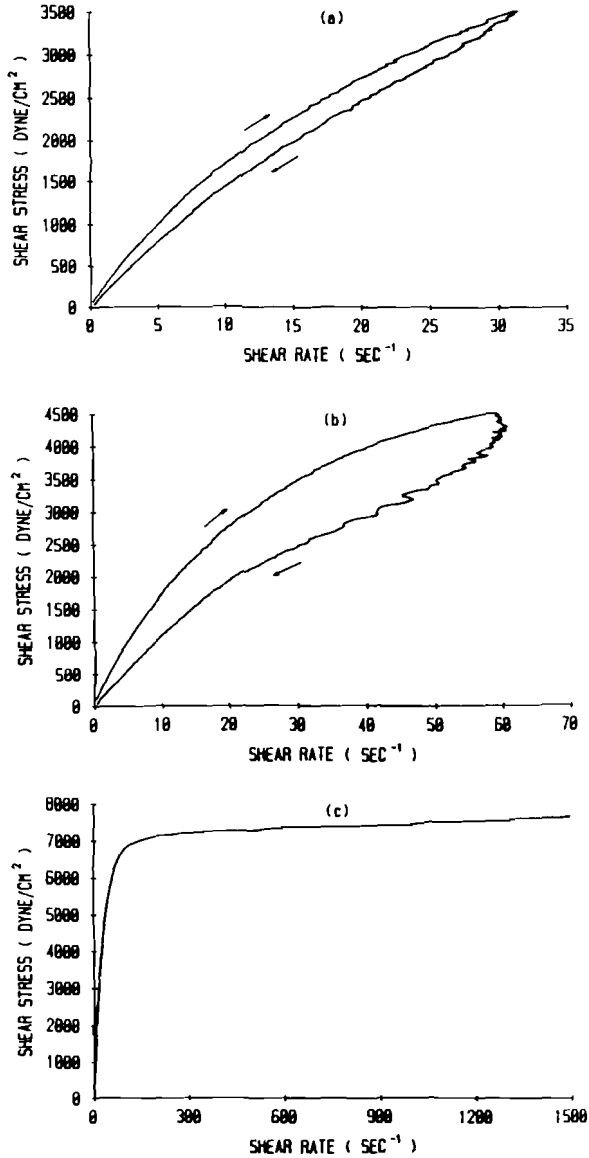


Figure 5. Effect of shear conditions on flow behavior of emulsion ink A containing 30% FS1. Programmed maximum shear stresses are (a) 3500, (b) 4500, and (c) 8000 dynes/cm². Samples were presheared at 5 sec⁻¹ for one minutes.

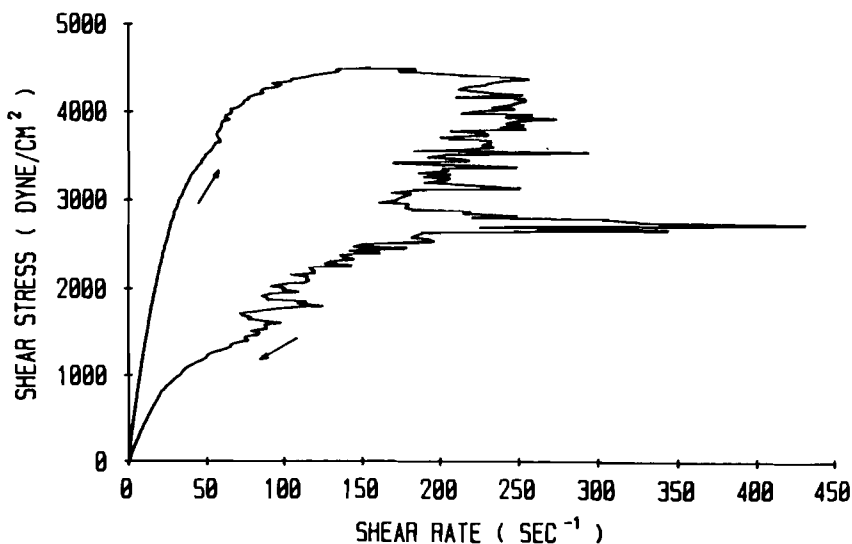


Figure 6. Effect of shear history on flow behavior of emulsion ink A containing 30% FS1. The sample was presheared at 30 sec^{-1} for 10 minutes.

The flow curves shown in Figures 5b and 6 reveal the effect of shear history on the stability of emulsion inks. Samples were respectively presheared at 5 sec^{-1} for 1 minute and at 30 sec^{-1} for 10 minutes before the flow measurements. Experimental conditions were otherwise the same for the two samples. Their flow curves differed significantly. Partial coalescence of emulsified fountain solution caused by the vigorous pretreatment temporarily destabilized the emulsion system.

Effect of Water Content

The effect of water content on the stability of emulsion inks under high shear conditions has been reported (Chou and Fadner, 1986), and briefly discussed in a preceding section. Figure 7 shows flow curves obtained under moderate shear conditions as a function of water content. Ink D containing 20% fountain solution FS2 appears slightly more stable than that with 30% water content, even though the measured maximum shear rate is higher. Emulsion breakdown occurs when the ink contains 40% fountain solution or more. The experimental results obtained under high and moderate shear conditions are consistent.

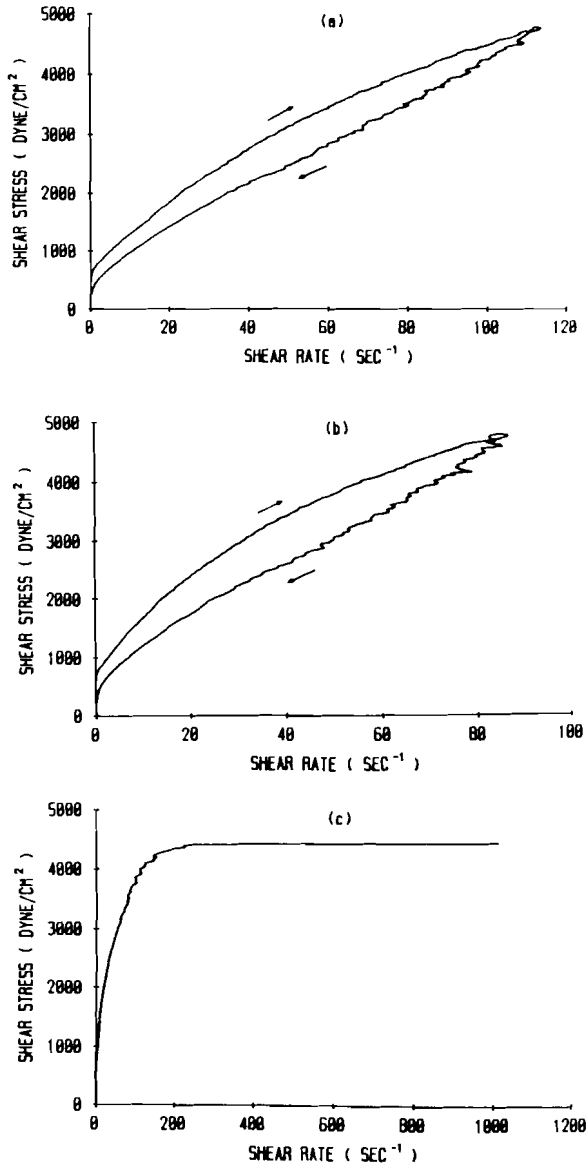


Figure 7. Effect of water content on flow behavior of emulsion ink D containing (a) 20%, (b) 30%, and (c) 40% FS2.

Effect of Temperature

Figure 8 displays typical examples of the viscosity versus temperature curves obtained with the controlled-rate rheometer for stable, metastable, and unstable emulsions. The results are consistent with those obtained from flow measurements.

Stable emulsion systems are identified by a smooth curve within the temperature range studied (Figure 8a). As expected, the viscosity increases exponentially with decreasing temperature. This type of curve is typical of all fresh inks.

The viscosity of a metastable emulsion ink also increases with decreasing temperature but reaches a maximum value, and the curve is somewhat irregular. The irregularity signifies concurrence of incipient water separation and emulsion reformation. As the temperature further decreases, the viscosity decreases rapidly but retains finite values (Figure 8b). This behavior indicates that emulsified fountain solution coalesces into discrete water layers.

The viscosity of emulsion ink B behaves similarly to the metastable system but drops to a value of about 0.4 poise at 27° C (Figure 8c), which is equivalent to that of fountain solutions. Subsequent decrease in the temperature does not change the viscosity of the system. This result evidences a gross separation of fountain solution from the ink. This catastrophic loss of viscosity characterizes unstable emulsion inks.

Corresponding results from the controlled-stress rheometer shown in Figure 9 illustrate the combined effect of temperature and shear stress on the stability of emulsion inks. Figure 9a shows a curve, obtained at an applied shear stress of 1500 dynes/cm², similar to that in Figure 8a. This emulsion was stable under the given set of conditions. An increase in the applied shear stress to 2500 dynes/cm² resulted in a curve typical of metastable emulsion inks (Figure 9b). The absence of a rapid reduction in viscosity, as shown in Figure 8b, is probably due to the different measuring systems discussed previously. Figure 9c shows that this emulsion breaks down at about 36° C when the applied shear stress is increased to 3800 dynes/cm². Both metastable and unstable systems exhibit an initial decrease rather than an increase in the viscosity as the temperature is lowered. Reasons for this unusual behavior are not known at present.

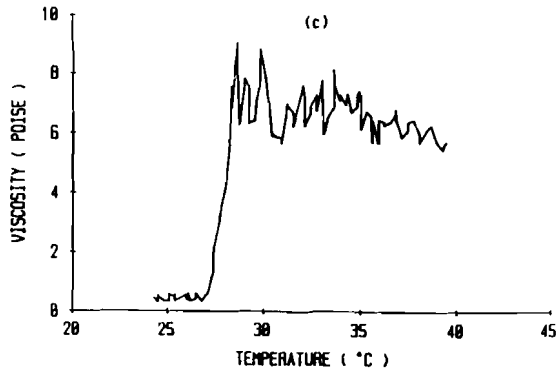
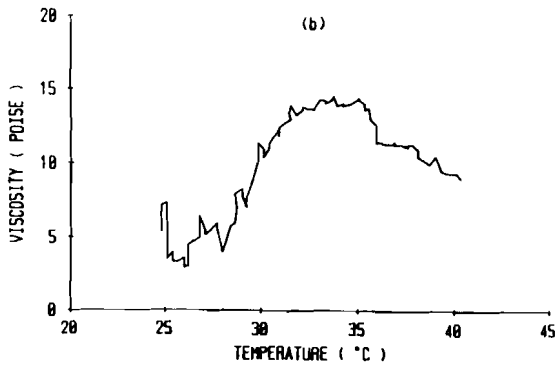
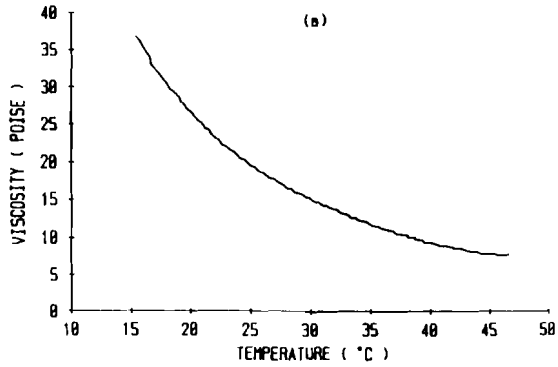


Figure 8. Viscosity versus temperature curves from the controlled-rate rheometry for (a) emulsion ink B with 40% FS1 at 80 sec^{-1} , (b) emulsion ink A with 30% FS1 at 100 sec^{-1} , and (c) emulsion ink C with 20% FS1 at 50 sec^{-1} .

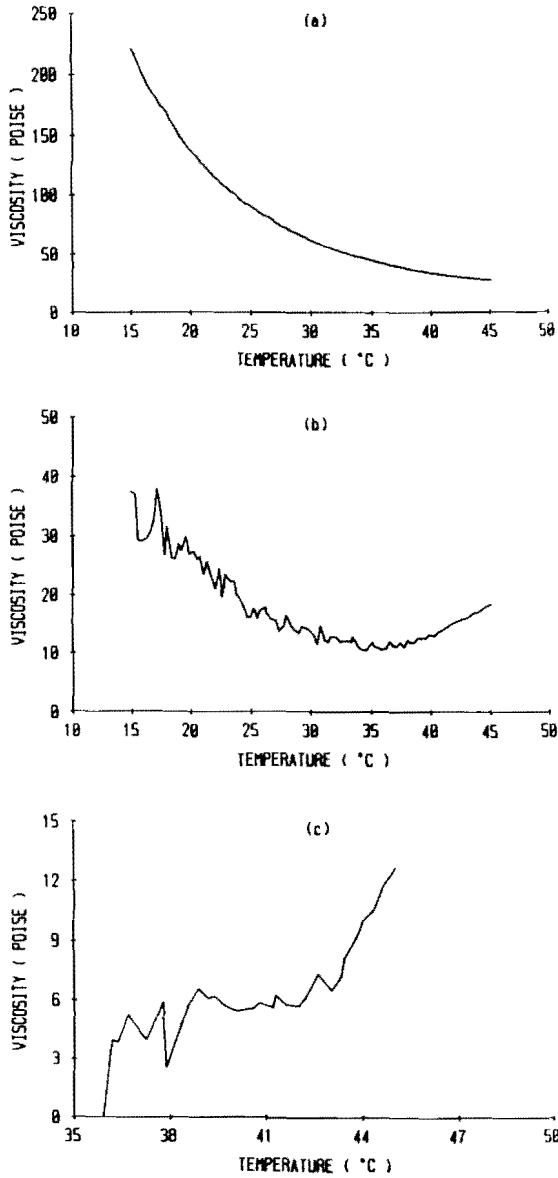


Figure 9. Viscosity versus temperature curves from the controlled-stress rheometry for the emulsion ink D with 20% FS1 at applied constant shear stresses of (a) 1500, (b) 2500, and (c) 3800 dynes/cm².

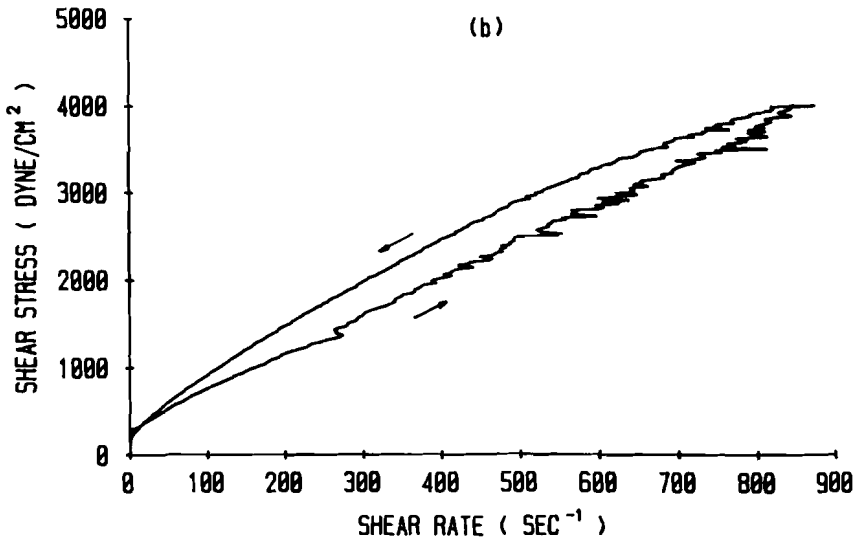
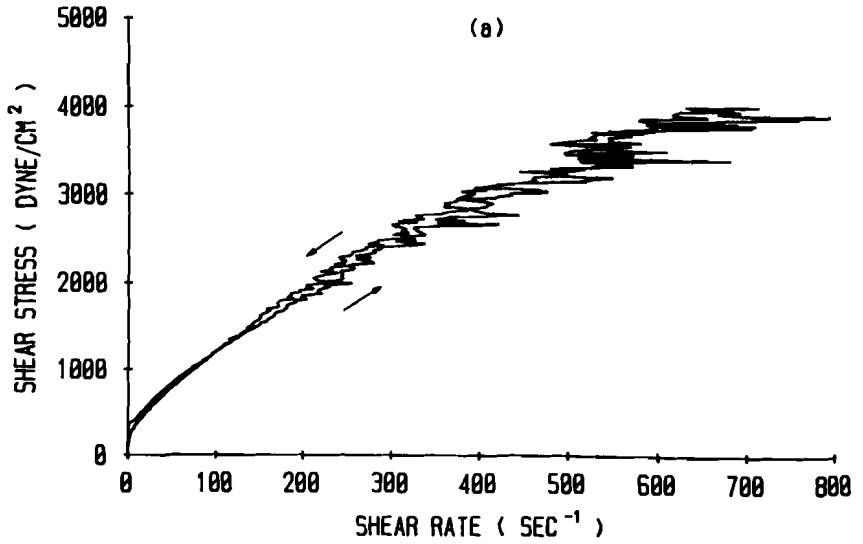


Figure 10. Effect of temperature on flow behavior of emulsion ink C with 20% FS1 at (a) 35° C and (b) 45° C.

Experimental results shown in Figure 10 verify that the flow measurement is also very useful for studying temperature effects. The emulsion ink C with 20% FS1 exhibited gross water separation at 25° C (Figure 4). It became metastable at 35° C (Figure 10a). Its flow curves at 45° C closely resemble those of stable emulsions (Figure 10b). However, the down curves appear above the up curves in both cases. This emulsion ink is rheopectic under the given sets of conditions.

The results from temperature sweep and flow techniques are consistent with our previous findings (Chou and Fadner, 1986). Emulsion inks are more stable at higher temperatures. A more precise discussion about the temperature effect on emulsion stability is not possible without knowing the identities of the surfactants in the inks and fountain solutions. However, as a general rule, surfactants become more soluble in the oil at higher temperature (Rosen, 1978), due to the decreased degree of hydration of the hydrophilic groups. This decreases the interfacial tension between the oil phase and the interfacial region. The interfacial tension between the water phase and the interfacial region increases correspondingly. The area between the interfacial region and the oil phase is greater than that between the interfacial region and the water phase in w/o emulsions. The total interfacial free energy can be reduced as a result of increasing temperature and the stability of emulsion thus increases.

Rheopectic Behavior of Emulsion Inks

Ink B with 40% and ink C with 20% of fountain solution FS1 exhibit rheopectic behavior (Figures 2 and 10). All the other emulsion inks we studied were thixotropic. The increasing viscosity with time associated with rheopectic behavior may be caused by the shear-induced buildup of internal structure or by the change in emulsion composition. The former is less likely because both emulsions exhibit shear thinning behavior.

Data from Karl Fischer titration showed a 4.6% loss of water content in the emulsion ink C containing 20% fountain solution FS1 as a result of the flow measurement at 45° C. This infers that evaporation of fountain solution during measurement can be the major reason for the apparent rheopectic behavior of this emulsion ink.

The time-dependent rheological property of a thixotropic fluid can be represented by a thixotropy index which is the

ratio of the hysteresis loop area to the area underneath the up curve (Chou and Bain, 1988). The loop area is calculated by subtracting the area underneath the down curve from that underneath the up curve. Figure 11 shows that the thixotropy index of emulsion ink B decreases with increasing water content. When the water content is greater than about 20%, the index value becomes negative. Similar results were observed when the same ink contained various quantities of fountain solution FS2.

The viscosities of inks B and C are so low that the activation energy for migration of emulsified fountain solution to the ink/air interface for subsequent evaporation is presumably low. Consequently, the evaporation rate is expected to increase with water content. This reasoning is consistent with the results shown in Figure 11; the emulsion ink containing more fountain solution is more rheopectic. Print tests of ink B on our keyless test press also showed that the same water buildup in the ink circulating system was observed whether the dampener was set at normal or unusually high input.

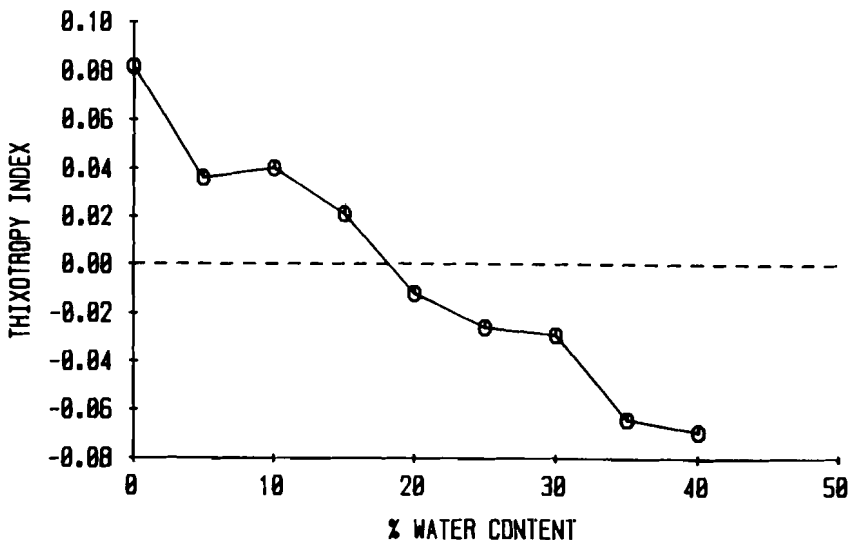


Figure 11. Thixotropy indices of emulsion ink B with FS1 as a function of water content.

CONCLUSIONS

1. Rheometry has proved to be a very useful technique for studying the shear stability of emulsion inks. The results from the cone-and-plate and concentric cylinder measuring systems are consistent, though they are quantitatively not the same for metastable and unstable emulsion systems.
2. Emulsion inks can be classified into stable, metastable, and unstable systems according to the shape of their flow curves and to the shape of their viscosity versus temperature curve.
3. A given emulsion ink may exhibit the characteristics of either a stable, a metastable, or an unstable system, depending on the shearing conditions. All emulsion inks are practically stable at relatively low shear rates. With increasingly vigorous shear conditions, emulsion inks become less stable and can eventually break down. The transition range from stable to unstable state for a given emulsion ink may be a function of the physico-chemical nature of the ink and fountain solution.
4. The stability of an emulsion ink decreases with increasing water content. Emulsion inks are more stable at higher temperatures.

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