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Particle motions in non-newtonian media

III. Further observations in elasticoviscous fluids *)

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With 3 figures and 2 tables

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1. Introduction

A recent paper from this laboratory has described in detail the translational and rotational motions of isolated, neutrally buoyant rigid rods (axis ratio $r_p \geq 10$) and discs ($r_p \simeq 0.1$) suspended in pseudoplastic and elasticoviscous fluids undergoing *Couette* flow (1). The deformation, orientation and burst of *Newton*ian drops in non-*Newton*ian media, and of non-*Newton*ian drops in *Newton*ian media, with the accompanying migration of the particles across the planes of shear were also studied (1).

This paper reports on some observations which have extended the previous work to rods and discs having particle axis ratios closer to unity, and to higher rates of shear. Experiments on the transient deformation and burst (2) of liquid drops in both *Newton*ian and elasticoviscous systems consisting of pairs of mutually soluble liquids having effectively zero interfacial tension, are also described.

Some of the results obtained are of a preliminary and qualitative nature; nevertheless they are of interest and considered to be worth recording since they show that certain of the particle motions are more complex than had previously been thought (1, 3).

2. Experimental part

The suspensions were contained in the annulus between counter-rotating cylinders of a *Couette* apparatus previously described (4, 5) and the particle motions viewed and photographed either along the X_1- or X_2-axes of the shear field (fig. 1–1a)[1]).

The systems studied are listed in table 1 and with the exception of aqueous solutions of guar gum (Jaguar A20D, Stein-Hall Ltd.), elasticoviscous and pseudoplastic solutions of polymers were prepared from materials previously used (1, 3). Their rheological properties were measured in a Mechanical Spectrometer (6, 7, Rheometrics Inc.) and the first normal stress differences (fig. 1–1b) and apparent viscosities are given in the table. The aqueous polyacryl-

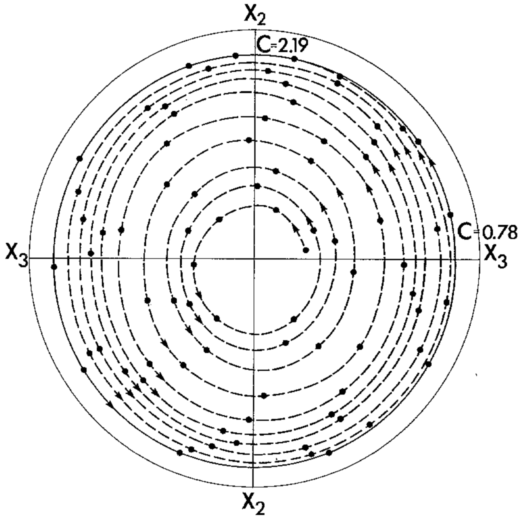

Fig. 1a. Measured projections of the semi-axis of revolution of a disc on the X_2X_3-plane; $r_p = 0.68$, system 1. The continuous change of the calculated conventional orbit constant with time is shown by the dashed line; the solid line shows the equilibrium orbit (with a periodically varying finite C) reached after 10 rotations, $G = 1.14\ \text{sec}^{-1}$. The outer circle corresponds to the orbit having $C = \infty$

*) This paper is, in effect, an addendum to Reference (1). To avoid unnecessary repetition, identical symbols have been used (with the addition of α' and D' which are defined in the text), and frequent use is made of figures and equations from references (1) and (2).

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[1]) Indicating fig. 1a of ref. (1).

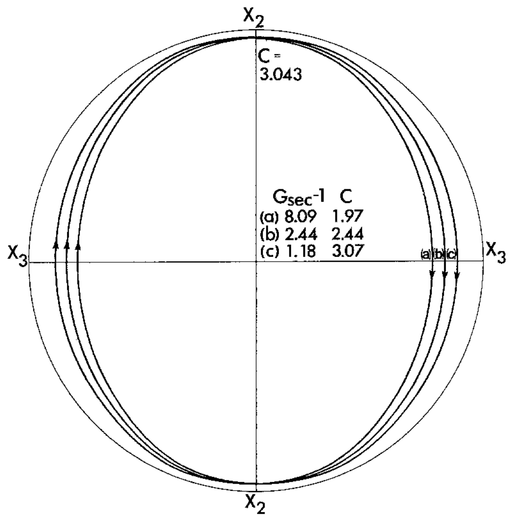

Fig. 1 b) X_2X_3-projections of the semi-axis of revolution of a disc, $r_p = 0.68$, $r_e = 0.63$ in system 1, rotating in equilibrium orbits at various shear rates. The outer circle corresponds to the orbit having $C = \infty$

amide solutions in systems 1 and 2, prepared under carefully controlled conditions using low stirrer speeds, exhibited normal stresses which at a given shear rate and polymer concentration were appreciably higher than those previously measured in a *Weissenberg* Rheogoniometer (1). This discrepancy is probably due to differences in the mode of solution of the polymer (6, 8, 9). Thus, in systems 6 to 9 in which 3.8% polyacrylamide solutions were prepared in a Waring blender, the apparent viscosities at a given G were lower than those in systems 1 and 2.

Discs of axis ratio 0.067 were prepared by punching out Mylar computer tape (10) and from these, particles having $r_p = 0.48$, 0.68 and 0.88 were made by stacking 7, 10 and 13 discs respectively in a jig using ethyl acetate as a solvent. Some swelling of the particles occurred during this process. Rods were machine cut from polyethylene filaments. Aluminum coated

Table 1. Physical properties of suspensions, 21.5 °C
a) Rigid rods and discs

System	Suspending phase	Apparent viscosity η_0 Poise		$p_{33} - p_{22}$, dyne cm^{-2}		Particles	Particle dimensions mm	
		$G = 1$ sec^{-1}	$G = 10$ sec^{-1}	$G = 1$ sec^{-1}	$G = 10$ sec^{-1}		2a	2b
1	2.5% polyacrylamide	68.0	21.0	153	672	Mylar discs	0.12 to 0.16	1.80
2	in water					Polyethylene rods	1.80 4.50	0.50 0.50
3	3% polyisobutylene	2.4	2.4	Not detectable		Polyester laminated AC discs	0.09	1.83
4	in Decalin					Aluminum coated nylon rods	3.00	0.18

b) Liquid drops

System	Suspending phase	Apparent viscosity, η_0[1]) Poise	Drop phase	Apparent viscosity, η_1[1]) Poise	λ[1])
5	Silicone (200 fluids)	21.4 104 200	Silicone (200 fluid)	200 55.0 55.0	9.3 0.50 0.28
6	Polyacrylamide 3.8% in water 5.0%	7.1 23.4	Polyacrylamide 3.8% in water 5.0%	23.4 7.1	3.3 0.30
7	3.8% polyacrylamide in water	7.9	1.5% Guar gum in water	42.6	5.4
8	3.8% polyacrylamide in water	7.9	5% polyisobutylene in decalin	9.8	1.2
9	3.8% polyacrylamide in water	7.1	0.05 to 0.25% Carbopol in water	2.1 to 11.3	0.3 to 1.6

[1]) For the non-*Newton*ian fluids, η_0 and η_1 are given at $G = 13$ sec^{-1} and $\lambda = \eta_1/\eta_0$ calculated from these values.

nylon rods and polyester laminated aluminum discs were used as conducting particles in experiments in which electrical fields were applied across the *Couette* cylinders.

Liquid drops were inserted from a hypodermic syringe needle placed at the radial position of the stationary layer. However, due to the appreciable lateral migration of the drops in the non-*Newton*ian liquids under the conditions of the experiments (1), movement of the particles out of the stationary layer soon occurred.

3. Results

a) Discs

(i) Period of rotation about X_1 (vorticity) axis

Previous observations made with discs of axis ratio between 0.06 and 0.18 in 2% aqueous polyacrylamide (1) had shown that below a critical rate of shear, the angular velocity $d\phi_1/dt$ of the axis of revolution of the discs was in accord with theory for *Newton*ian fluids, as given by eq. [1–11] [2]) using an equivalent ellipsoidal axis ratio r_e determined from the particle period of rotation T in a *Newton*ian medium. Above the critical G (1) the discs rotated until their faces became oriented in the direction of flow, $\phi_1 = 0$, $\theta_1 = \pi/2$, and ceased to rotate further.

Here, however, with discs of axis ratios 0.48 to 0.88 in system 1, $d\phi_1/dt$ was found to be lower than predicted by the theory and the particles took appreciably longer to execute a complete orbit. This is shown in table 2 which lists observed and calculated values of T, the latter being obtained from eq. [1–12] using r_e computed from the empirical relation

$$\log_{10} r_e = 0.78 \log_{10} r_p + 0.051\,, \qquad [1]$$

which is valid in the range $0.1 \leq r_p \leq 50$ (11). It is evident from the table that at a given r_p, the ratio T_{obs}/T_{calc} increased with increasing G, while increasing r_p at a given G did not affect the ratio significantly.

(ii) Drift in orbit constant

The observed periods of rotation of thin discs, $r_p = 0.067$, at *low* G ($<0.15\ \text{sec}^{-1}$) were also greater than those calculated (table 2), and unlike previous results in which the particles were found to drift into orbits of $C = \infty$ (fig. 1–1c), here the discs drifted into equilibrium orbits in which $C < \infty$ and periodically varied between a maximum at $\phi_1 = 0, \pi$ and a minimum at $\phi_1 = \pi/2, 3\pi/2$.

Discs of larger axis ratio also showed this behaviour at higher G, as is illustrated in fig. 1a by a plot of the X_2X_3-projection of the axis of revolution (the semi-minor axis, eq. [1–14]) of a disc, $r_p = 0.68$, in which the orbit constant at a given ϕ_1 continually increased, starting at an initial value close to $C = 0$. As the shear rate was further raised, the value of C at $\phi_1 = \pi/2$ decreased while that at $\phi_1 = 0$ remained sensibly constant (fig. 1b). It should be noted that although the discs never rotated in a truly spherical elliptical orbit with a constant C, the instantaneous values of the orbit constant were computed from eq. [1–13] using the measured angles ϕ_1 and θ_1 and r_e determined from eq. [1]. At $r_p = 0.067$, however, increasing the shear rate resulted in the particle drifting into the limiting orbit $C = \infty$, as found before (1) and at sufficiently high G aligning itself with the flow without further rotating.

b) Rods

The gradual, steady drift in orbit of rods to an orientation where the major axis is aligned with

[2]) Indicating eq. [11] of ref. (1).

Table 2. Effect of shear rate and axis ratio on period of rotation of discs in 2.5% polyacrylamide solution

r_p	r_e	G sec^{-1}	T sec obs.	T sec calc.[1])	$T_{obs.}/T_{calc.}$
0.067	0.141	0.15	490	303	1.62
0.477	0.634	0.15	96.0	93.0	1.04
		1.14	16.4	12.2	1.35
		1.18	16.8	11.8	1.43
		2.44	9.70	5.69	1.70
		3.47	7.32	4.00	1.83
		5.40	5.55	2.57	2.15
		6.53	5.10	2.13	2.40
		8.09	4.20	1.72	2.45
		8.77	4.10	1.58	2.59
		10.6	3.94	1.31	3.02
0.675	0.832	0.15	97.0	85.0	1.14
		1.14	14.8	11.2	1.32
		3.00	5.60	4.26	1.32
		5.40	4.80	2.37	2.03
0.881	0.905	0.15	90.0	84.6	1.07
		1.14	12.9	11.1	1.17
		3.00	5.80	4.21	1.32
		5.40	4.65	2.34	1.99

[1]) From eq. [1–12] using the measured G and calculated r_e.

the vorticity axis, $C = 0$ (fig. 1–1c, 1, 3) was again observed in this study at shear rates $< 5\ \text{sec}^{-1}$ in system 2. However, at higher shear rates a behavior analogous to that of the non-rotating disc with its faces aligned with the direction of flow was found with rods, $r_p = 9.0$. This occurred when the rod was initially aligned with its major axis along the X_2-axis and lay in, or very close to, the X_2X_3-plane ($\theta_1 = \pi/2$). Upon shearing the fluid, the particle rotated to the orientation $\phi_1 = \pi/2$ and then, once aligned with the flow, ceased to rotate. The rods remained in this orientation provided that there were no particle interactions or disturbances to the flow. If these occurred, the particles resumed their angular motion and drifted into the orbit $C = 0$.

A variation of the motion described above was found with the rods of lower $r_p (= 5.6)$ which if initially placed in the orientation $\phi_1 = 0$ in the X_2X_3-plane also rotated through the first quadrant of their orbit with only small changes in orbit constant and became aligned with the X_3-axis. There was then no further change in ϕ_1- or θ_1-orientation for a period as long as 10 sec and during this "residence" time the rod migrated slowly towards the outer cylinder wall (1). After this interval there followed a shorter "induction period" during which the

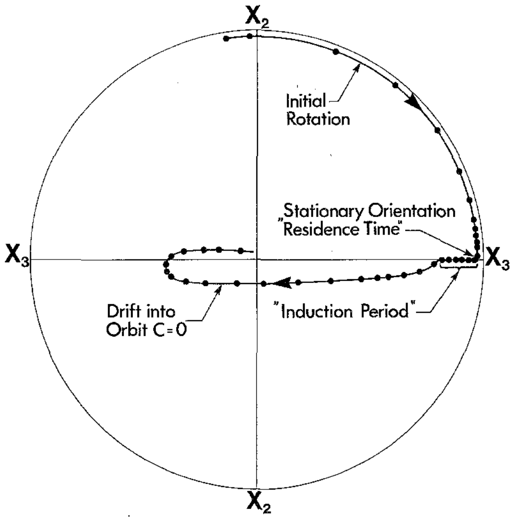

Fig. 2. Measured projections of the semi-major axis of a rod on the X_2X_3-plane, $r_p = 5.6$, $G = 6.43\ \text{sec}^{-1}$, system 2, as the particle first becomes oriented with its major axis in the direction of flow before finally drifting into the orbit, $C = 0$, in which the major axis is aligned with the X_1 (vorticity)-axis. The experimental points were recorded at 1 min intervals; the outer circle corresponds to the orbit having $C = \infty$

angle θ_1 decreased without a change in the angle ϕ_1. This continued until the rod reached a critical θ_1-orientation when the angular rotation of the rod resumed and the particle drifted through a series of orbits to a final orientation $C = 0$. Fig. 2 illustrates the four consecutive stages of this behavior.

c) Rods and discs in shear and electric fields

Single, isolated electrically conducting rods and discs in systems 3 and 4 were subjected to *Couette* flow until the equilibrium orbits of $C = 0$ and $C = \infty$ respectively had been reached. A 60 Hz electric field of about 400 volts/cm was then applied across the annular gap of a *Couette* having stainless steel cylinders electrically insulated from each other (4). It was observed that both discs (whether non-rotating or rotating) and rods drifted from the limiting orbits into equilibrium orbits in which $0 < C < \infty$, the value of C decreasing (discs) or increasing (rods) as the magnitude of the [sub-critical (12)] electric field was increased. When the electric field was removed, the original limiting orbits were regained. Similar behaviour was observed with these particles in a 0.25% pseudoplastic solution of Carbopol in propylene glycol (13). This illustrates that simultaneous application of a shear field (tending to cause an orbital drift in a non-*Newton*ian medium in one direction) and a sub-critical electrical field along the X_2-axis (tending to cause drift in the opposite direction (14)) leads to an intermediate equilibrium orbit $0 < C < \infty$ in which the two opposing effects become balanced.

d) Transient deformation and burst of drops, $\gamma = 0$

It has been shown (2, 15) that a drop suspended in a fluid which is suddenly sheared from rest will undergo a periodic deformation which appears as a "wobble". The instantaneous deformation D' of the drop into an ellipsoid and its orientation α' with respect to the X_2-axis of the flow field (fig. 2–1) undergo a damped oscillation. In the special case of two mutually soluble liquids ($\gamma = 0$) considered here, D' and α' oscillate without damping with a period $T_0 = 2\pi/G$ when the drop is much more viscous than the medium. When $\lambda \leq 1$, however, there is no wobble; the drop is then continuously extended and aligned with the flow as D' and α' tend monotonically to 1 and 90°, respectively (fig. 2–2).

(i) Newtonian drops

A typical result obtained in silicone oil (system 5) for $\lambda = 9.3$ is shown in fig. 3. Little or no damping of the wobble was observed, but as can be seen the measured $T_0 > 2\pi/G$, and D' was greater than the value predicted by theory (eq. [2–15a]). The orientation α' increased from an initial value of 45° to 135° as predicted, but at a rate which fluctuated somewhat. Although the measured T_0 were always greater than the theoretical values, the transient deformations exhibited a scatter within the same system, being mostly greater but occasionally also smaller than the predicted values. This result probably reflected an error inherent in the experimental method due to the difficulty of producing a spherical drop in a system of zero interfacial tension. It was thus likely that one sometimes observed a time dependent deformation D' superimposed on that of an initially non-spherical drop.

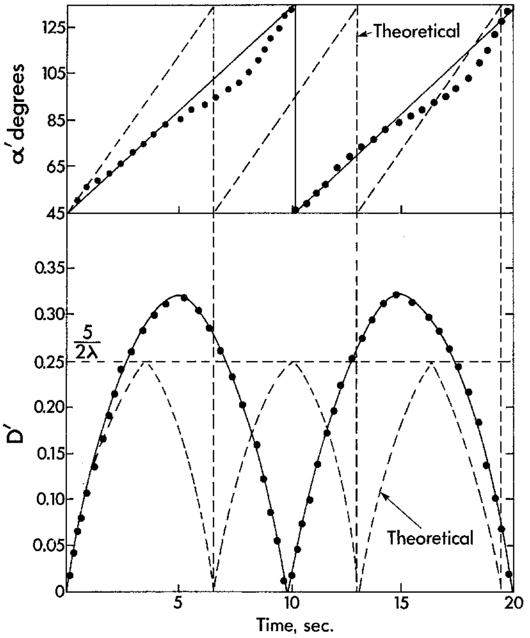

Fig. 3. The undamped oscillation (wobble) of the deformation D' and orientation angle α' (c.f. fig. 2–1) of a soluble 200 poise silicone oil drop in a 21 poise silicone oil, $G = 0.95\ \text{sec}^{-1}$. The points are experimental and the dashed lines are calculated from eqs. [2–15a] and [2–15b] with a period of oscillation $T_0 = 2\pi/G = 6.45$ sec

At viscosity ratios $\lambda = 1/4$ and $1/2$, the transient deformation was found to increase from 0 to 1 as the particle became continuously extended and totally aligned with the flow ($\alpha' = 90°$). Both D' and α' were greater than the values predicted by theory, eqs. [2–32] and [2–33].

(ii) Elasticoviscous drops

When viewed along the X_1-axis, the deformation and burst of a viscous polyacrylamide drop suspended in a less viscous polyacrylamide medium was complex. A somewhat clearer picture emerged when the events were viewed in a direction normal to the planes of shear, i.e., along the X_2-axis. The initially spherical drop became extended *vertically* (i.e., along the vorticity-axis X_1) into a cylindrical shape. This then exhibited a rocking motion from side to side about the vorticity-axis, somewhat reminiscent of the rotation of a rigid rod in a spherical elliptical orbit in *Couette* flow (16, 17). As the longitudinal deformation continued a buckling motion set in, which resulted in the formation of thin threads which finally broke up into discrete cylindrical daughter drops.

Although the time to burst in a given system decreased with increasing shear rate, the mode of extension of the drop and final burst were identical even at G as low as $0.05\ \text{sec}^{-1}$. Moreover, when the suspension continued to be sheared after break-up, the daughter droplets in turn exhibited the same longitudinal deformation into cylinders and buckling, thus resulting in a large number of very small cylindrical droplets which eventually became dissolved in the suspending phase.

Similar behaviour was observed with the guar gum drops in system 7, except that here the cylinders became varicose and necked off at intervals along their length as the deformation continued, possibly because of the existence of a finite interfacial tension.

The effect of interfacial tension on the mode of drop deformation and burst in an elasticoviscous system was also explored using drops of polyisobutylene in Decalin suspended in aqueous polyacrylamide (system 8, $\lambda \simeq 2$ at $G = 1\ \text{sec}^{-1}$) in which it was visually apparent that γ was appreciable. It was observed that the deformation and burst over a range of G from 0.05 to 5 sec^{-1} occurred in a manner previously described as class BI (18), i.e., when shear was suddenly imposed, the drop extended in the X_2X_3 plane into an ellipsoid at low G; at higher G the central portion was pulled out into a cylinder in the X_2X_3 plane which above a certain critical value of the gradient necked off in the middle and formed two large droplets separated by tiny satelite droplets. The same progressive deformation and burst occurred under conditions of equilibrium deformation when G was increased slowly.

At viscosity ratios $\lambda < 1$ deformation of drops in system 5 and 6 was similar to that described above in the *Newton*ian silicone oil.

(iii) Pseudoplastic drop in elasticoviscous medium

The deformation and burst of aqueous Carbopol drops in system 9 was found to be independent of the viscosity ratio: when suddenly sheared, the initially spherical drop deformed into an ellipsoid from which *continuous* threads of liquid were withdrawn from the pointed ends in the X_2X_3 plane. Thus, the drop, while retaining its ellipsoidal shape, gradually diminished in size with time. The continuous ejection of a thread, rather than that of discrete droplets as in type A break-up previously found in *Newton*ian systems (18, 19) as well as for pseudoplastic drops in *Newton*ian media (1), is possibly associated with the low value of the interfacial tension in the systems used here.

Concluding remarks

The previous study (1) with rods and discs in pseudoplastic Carbopol solutions undergoing *Couette* flow had shown good agreement between the experimental and calculated periods of rotation, similar results being obtained in elasticoviscous aqueous solutions of 3% polyacrylamide, providing the shear rate did not exceed a critical value. The present investigation has demonstrated that even at relatively low shear rates in 2.5% polyacrylamide solution there is a pronounced increase in the period of rotation from that predicted by theory, and that this difference further increases with increasing shear rate. This effect is most likely due to the existence of an elastic restoring torque, opposing that due to viscous deformation of the fluid. At low G it results in the retardation of the particle angular velocity and eventually, when the shear rate reaches a critical value, in cessation of particle rotation after the faces of the discs become oriented in the direction of flow. For the first time also, an alignment of rods in the direction of flow was observed, although here the equilibrium position in which the particle lies in the X_2X_3-plane is a metastable one.

That some of these phenomena were not previously observed may in part be a reflection of the much higher normal stresses and, one pressumes, elastic moduli (6) of the polyacrylamide solutions prepared for system 1. That drift into limiting rotational orbits occurs in both pseudoplastic and elasticoviscous polymer solutions (1), whereas the impedance of particle rotation is only observed in elasticoviscous media suggests that it is the first normal stress difference which may be responsible for orbital drift. It is possible also that the somewhat surprising deformation of the elasticoviscous drops in system 5 ($\lambda > 1$) along the vorticity axis, rather than in the X_2X_3-planes as observed in *Newton*ian systems (18, 19) occurs as a result of the action of the same stresses which bring about the drift of rigid rods into the orbit $C = 0$.

More definitive explanations of the observed particle behaviour, however, must await the results of further experiments and the development of theory applicable to non-*Newton*ian fluids.

Summary

The rotation of rigid rods and discs, and the deformation and burst of liquid drops were observed in elasticoviscous solutions of polyacrylamide undergoing *Couette* flow at shear rates from 0.1 to $\sim 10\ \text{sec}^{-1}$. The study extended previous work (1) to cylindrical particles having axis ratios closer to unity and to liquid drops in systems having zero interfacial tension.

The period of rotation of discs was appreciably longer than predicted by theory applicable to *Newton*ian fluids. Depending on the axis ratio and shear rate, the particles drifted into orbits in which the conventional orbit constant C varied periodically throughout each orbit. Above a given shear rate, rods aligned themselves in the direction of flow without rotating, in a metastable equilibrium position; in the case of discs at higher shear rates the position of alignment was stable.

The oscillating deformation and orientation of drops in a system of two soluble *Newton*ian silicone oils was in fair accord with theory. With pairs of elasticoviscous polyacrylamide solutions, deformation and burst of viscous drops occurred along the vorticity axis, in contrast to the behaviour of elasticoviscous drops of polyisobutylene in Decalin having a finite interfacial tension, which deformed and burst in a manner similar to *Newton*ian drops.

Zusammenfassung

Die Rotation starrer Stäbchen und Scheibchen sowie die Verformung und das Aufbrechen flüssiger Tröpfchen wurde in viskoelastischen Polyacrylamid-Lösungen untersucht, die in einer *Couette*-Apparatur mit Schergeschwindigkeiten zwischen 0,1 und ca. 10 sec^{-1} geschert wurden. Diese Untersuchung ergänzt eine frühere Arbeit (1), insofern zylindrische Teilchen mit einem Achsenverhältnis näher bei eins und Tröpfchen in Systemen mit verschwindender Grenzflächenspannung berücksichtigt werden.

Die Umdrehungsperiode der Scheibchen kam erheblich langsamer heraus, als sie auf Grund der für *newton*sche Flüssigkeiten gültigen Theorie vorausgesagt wird. Die Teilchen trieben, abhängig von ihren Achsenverhältnissen und der Schergeschwindigkeit, in Kreisbahnen hinein, für welche die konventionelle Kreisbahnkonstante C sich periodisch änderte. Oberhalb einer bestimmten Schergeschwindigkeit orientierten sich die Stäbchen in Richtung der Strömung in einer metastabilen Gleichgewichtseinstellung; für Scheibchen war bei höheren Schergeschwindigkeiten diese Einstellung stabil.

Oszillationsverformung und Orientierung der Tröpfchen in einem System zweier ineinander lösbarer *newton*scher Siliconöle stimmte zufriedenstellend mit der Theorie überein. Für Paare viskoelastischer Polyacrylamid-Lösungen wurden die zähen Tröpfchen entlang der Richtung der Rotationsachse verformt und aufgebrochen im Gegensatz zu viskoelastischen, eine endliche Grenzflächenspannung aufweisenden Tröpfchen von Polyisobutylen in Dekalin, die ähnlich wie *newton*sche Tröpfchen verformt und aufgebrochen wurden.

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