

# Separation of Particles of Different Sizes from Non-Newtonian Suspension by Using Branched Capillaries

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In this experimental study, we have investigated the separation of particles with different sizes dispersed in liquids using the flow through branched capillaries. We investigated four different kinds of fluids of a Newtonian fluid, shear thinning fluid, non-shear thinning elastic fluid and shear thinning and elastic fluid. Only in the shear thinning fluid, the separation of large particles from a mixture of particles with different sizes was achieved. This separation has been found to be possible because of the fast migration of non-interacting particles across streamlines and the proper partitioning characteristics at the branch in shear thinning fluids. From the experimental results it has been concluded that the scale down of the system to miniaturize the system will be possible and therefore the process intensification can be achieved.

## Introduction

Separation of particles from the carrier liquid of suspension has been an important issue in chemical engineering. Recently more attention has been paid to the separation of different size particles suspended in very small amount of liquids of various kinds. For example, in a micro-TAS (Total Analysis System), one drop of human blood is used and one needs to separate red blood cells (RBC) from the mixture of white blood cells, platelets and RBC suspended in the non-Newtonian solution of many different kinds of protein. Recently Roberts and Olbricht (2003) have reported that with branched capillaries, particles can be separated from the liquid efficiently. Especially they have reported that more particles move to the branched capillaries depending upon the angle of the branch and the ratio of the flow to the branch, hence the separation characteristics changes. Their pioneering report may be utilized for many systems with different purposes. Following this, we have investigated the possibility by using these ideas of separating particles from particle mixtures suspended in liquids. To do so, we first reviewed the theory of migration briefly, which is the basis of particle separation.

When the suspending fluid is a Newtonian fluid, it has been well documented that, in the laminar regime, a single particle moves toward the midway be-

tween the center and the wall of channel due to the inertia of fluid when the particle loading is small. This is known as the Segre-Silberberg effect (Segre and Silberberg, 1961; Ho and Leal, 1974). This inertial migration mechanism can be utilized to separate particles (Poflee *et al.*, 1997, 1998), but Roberts and Olbricht (2003) pointed out that the inertial migration would be too slow a process to use for small particles because the migration rate scales with the particle Reynolds number. Therefore when particle Reynolds number is vanishingly small, a single particle will not move across the streamline in a tube flow in a finite time span. In the case of shear thinning fluids, Karnis and Mason (1966) first reported that a single particle moves toward the wall and Huang and Joseph (2000) later confirmed it numerically. In a viscoelastic fluid it was reported that a single particle moves toward the tube center (Karnis and Mason, 1966; Gauthier *et al.*, 1971). In an Oldroyd-B fluid which is a non-shear thinning viscoelastic fluid, a single particle tends to move either towards the wall or towards the center depending on the particle-tube diameter ratio (Huang *et al.*, 1997). The observed migration phenomena in viscoelastic and shear thinning liquids can be explained qualitatively as follows: Let us consider a spherical particle suspended in a flowing liquid through a tube. In the shear flow field, the sphere will rotate according to the Jeffery orbit. Since the flow is non-homogeneous, i.e., the shear rate is not spatially the same, the normal stress distribution is not symmetrical with respect to the plane intersecting the sphere center. In the case of non-Newtonian fluid with non-zero normal stress differences, the imbalance of force acting on the sphere results in the migration of the sphere either to

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the center or to the wall. While the sphere surface facing the centerline is moving in the same direction as the main fluid flow, the sphere surface facing the tube wall is moving in the opposite direction to the main fluid flow so that shear rate is much higher than the other side. Therefore, when the normal stress difference coefficient is not shear thinning strongly, the normal force acting on the surface facing the centerline will be smaller than the force acting on the surface facing the wall. This will result in the migration in the direction of the tube center in the case of viscoelastic fluid. In the case of shear thinning fluid the normal stress difference coefficient is strongly shear thinning so that the normal force acting on the surface facing the tube wall is not large enough to overcome the normal force on the other side of the sphere and this will result in the migration in the opposite direction. More quantitative distribution of stress in an Oldroyd-B fluid or shear thinning fluid can be found in Huang and Joseph (2000).

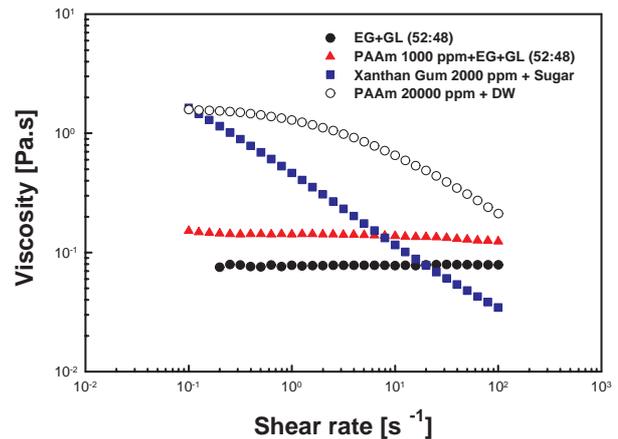
In a non-shear thinning elastic fluid, Kim (2001) has reported that the particles in polymeric suspension move toward the center due to the strong particle-particle interaction even when the particle loading is very small and the polymer concentration is very low, hence the elastic and shear thinning effects are very small. On the other hand in the flow of highly shear thinning fluid flow, the particles tend to align and then chain, and become a big aggregate (Tehrani, 1996). The results by Karnis and Mason (1966), Gauthier *et al.* (1971), Kim (2001) and Tehrani (1996) suggest that if we vary the rheological properties of carrier fluid judiciously we can control the direction of particle movement in branched capillaries with different flow rate. But there should be a process window for such an operation which may be quite narrow.

In this research we have extend Roberts and Olbricht's idea (2003) to the system with a distribution of particle sizes. We have investigated whether we can separate small or large particles from the mixture of particles with different sizes by using non-Newtonian carrier liquids to facilitate the separation while using the same geometry. This new idea can be utilized in the miniaturization and hence the process intensification ultimately.

## 1. Experimental

### 1.1 Materials

As the spherical particles we used PMMA particles. By sieving with a set of standard sieves, we collected particles with the diameters of  $70 \pm 15$  and  $200 \pm 10 \mu\text{m}$ . For easier visualization, the smaller particles were dyed in black while the larger particles were dyed in red using disperse dyes. As carrier liquids, we have chosen four different kinds of liquids: Newtonian fluid, non-shear thinning elastic fluid, strongly shear-



**Fig. 1** Viscosities of Newtonian fluid and polymer solutions: Polyacrylamide (PAAm) solution in the mixture of ethylene glycol and glycerin shows almost non-shear thinning characteristics. PAAm solution in distilled water shows shear thinning when the shear rate exceeds  $1 \text{ s}^{-1}$ . Xanthan gum (XG) in a sugar solution is severely shear thinning

thinning but not strongly elastic fluid and both shear thinning and elastic fluid. As the Newtonian fluid 52:48 mixture of ethylene glycol and glycerin was used. This fluid has the same density as the PMMA-particle ( $\rho = 1,188 \text{ kg}\cdot\text{m}^{-3}$ ), hence the sedimentation effect was eliminated. To investigate the effect of elasticity, a small amount (1,000 ppm) of polyacrylamide (PAAm, Aldrich Chem. Co., MW = 5,000,000 Da) was added to the Newtonian fluid. As a shear thinning solution, 2000 ppm solutions of xanthan gum (GR Grade, Sigma-Aldrich Inc.) in aqueous solutions of sucrose were used. The weight percent of sucrose was 50% to match the density. Finally an aqueous solution of polyacrylamide was prepared. The concentration of aqueous solutions was 2 wt%. Since the aqueous solution has large viscosity as shown below, we assumed that density matching was not required. This solution shows both shear thinning and viscoelastic properties. Both PAAm and xanthan gum were used as received. The rheological properties of the polymer solution were measured using the Couette or cone-and-plate fixtures of ARES-LS from Rheometrics Inc. In **Figure 1**, the viscosity is plotted against the shear rate for carrier liquids tested in this study. The aqueous solution of polyacrylamide exhibits the typical behavior of Boger Fluid: For more than two decades of shear rate from  $0.1 \text{ s}^{-1}$ , the viscosity of 1,000 ppm solution shows almost no shear thinning and the power law index is 0.94 in the shear thinning range. The xanthan gum solution is extremely shear thinning and the power law index is 0.44 between  $0.1$  and  $100 \text{ s}^{-1}$ . The aqueous solution of polyacrylamide has a zero-shear viscosity when the shear rate is less than  $1 \text{ s}^{-1}$  while it has a shear thinning region with the power law index of 0.84 when the shear rate is larger

than  $10 \text{ s}^{-1}$ . The linear viscoelastic properties of xanthan gum solutions and polyacrylamide solutions are also measured and plotted in **Figure 2**. It has been found that the 1000 ppm solution in the ethylene glycol and glycerin mixture is only weakly elastic. We also have listed the fluids and their rheological properties in **Table 1**.

### 1.2 Apparatus

In this research we set up a branched capillary system as shown in **Figure 3**. The apparatus consisted of a T-shaped flow cell, micro-syringe pump, two reservoirs and needle valves for flow rate control. The flow cell was constructed from a PMMA block of  $50 \text{ mm} \times 30 \text{ mm} \times 20 \text{ mm}$  by drilling capillaries using precision drills of 1 mm in diameter. The three branches are in the same plane. The length of the inlet region is 15 mm and the length of the outlet is 10 mm. The cell was placed in such a way that the plane made of the T-shaped branch is perpendicular to the gravity. Particle laden fluid was pumped to the cell by using a syringe pump of 50 ml (KDS-230, KD-Scientific). The fluid coming out of each branch was sent to the waste reservoir. Next to the reservoir was a needle valve to control the flow rate. A mesh screen was installed within the reservoir not to get particles clogged within the valve. Between two elements of any kind, plastic tubing (1/4" Tygon tube) was used. Flow rate was determined by measuring the amount of effluent from the metering valve. Using a digital camcorder (DCR-VX2000, SONY Corp.) each experimental run was recorded. The number of particles to each branch was counted by eyes while examining the recorded experimental runs.

## 2. Results and Discussion

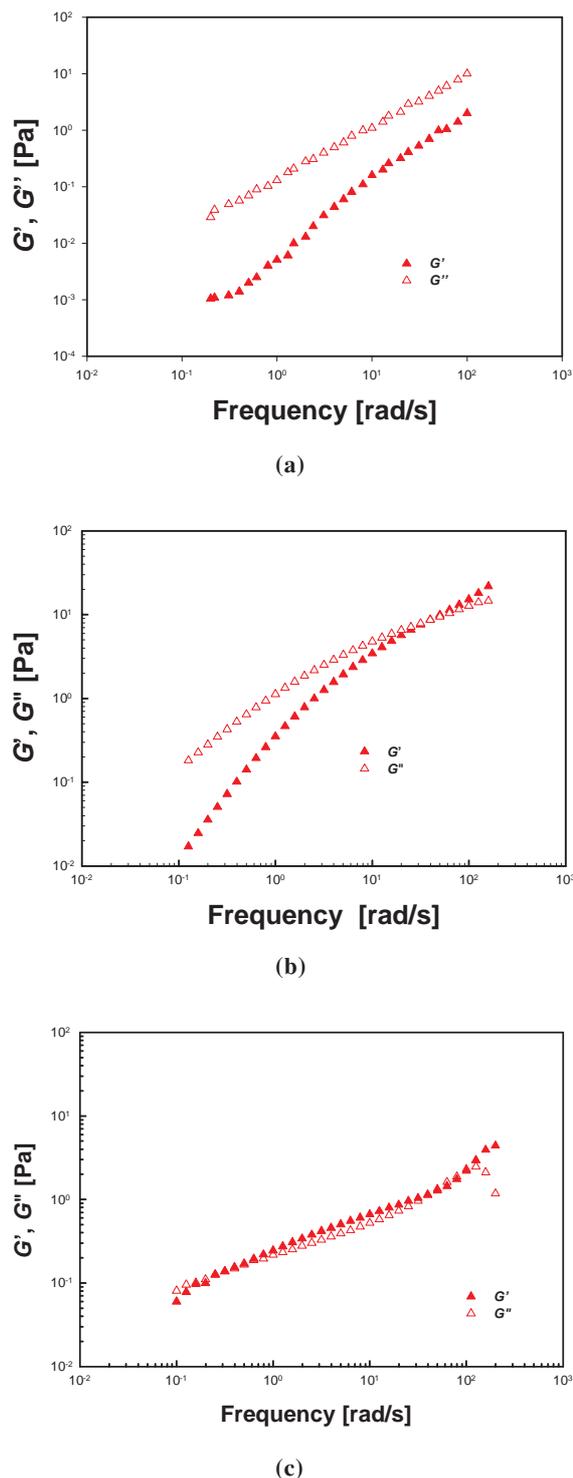
Depending on the ratio of the flow through the straight tube to the total flow rate, the number of particles changes that go with the fluid. In **Figure 4**, we define the relevant variables. The mass balances for both the fluid flow and the number of particles are

$$Q_T = Q_1 + Q_2 \quad (1)$$

$$N_T = N_1 + N_2 \quad (2)$$

where  $Q$  and  $N$  denote the flow rate and the number of particles passing a capillary in a unit time, respectively. We report the data for the separation of particles at the branched capillaries by plotting  $N_1/N_T$  as a function of  $Q_1/Q_T$  as used by Roberts and Olbricht (2003).

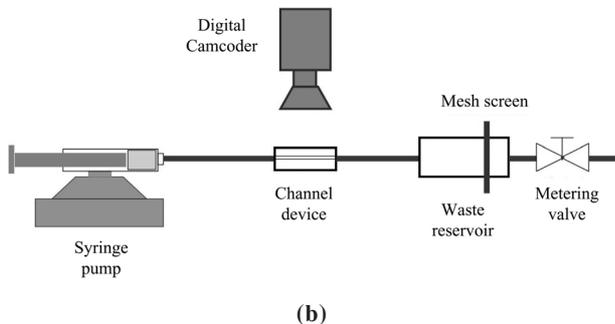
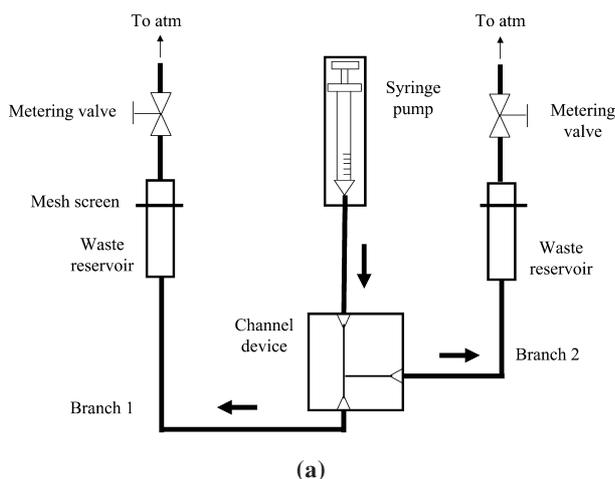
Experimental results for the Newtonian fluid are shown in **Figure 5**. Here we added 1% of the larger particles and 1% of the smaller particles in the liquid. Hence the total volume fraction of solid was 2%. We kept this fraction throughout this research except for



**Fig. 2** Viscoelastic properties of dispersing fluids: (a) 1000 ppm polyacrylamide solution in ethylene glycol and glycerin mixture; (b) aqueous solution of 2% polyacrylamide; (c) 2000 ppm xanthan gum solution in aqueous solution of 50% sugar. In (a) the solution shows a very weak elasticity.  $G'$  values for frequency values smaller than  $3 \text{ s}^{-1}$  in (a) are meaningless since they were collected beyond the precision limit of rheometer. The slope of  $G'$  should be 2 in the dilute limit

**Table 1** Rheological properties of fluids treated in this research

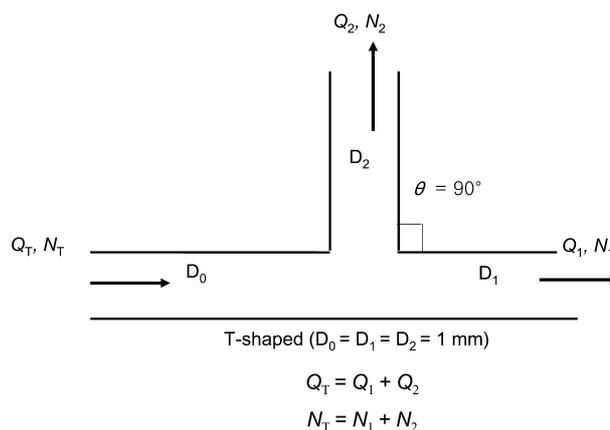
	$\eta_0$ [Pa·s]	$n$	Power law range [s <sup>-1</sup> ]	Characteristic time, $\lambda$ [s]	Description
Ethylene glycol (52%) + glycerin (48%)	0.075	1	All values	0	Newtonian
1000 ppm polyacrylamide + above solution	0.136	0.94	10–100	1.28	Non-shear thinning elastic
2000 ppm xanthan gum solution in 1:1 water–sugar mixture	N.A.	0.44	0.1–100	Not measured	Shear thinning
2% aqueous solution of polyacrylamide	1.57	0.84	8–100	5.43	Shear thinning and elastic



**Fig. 3** Schematic diagram of the experimental apparatus: (a) top view; (b) side view

the last case when we investigated the effect of particle–particle interaction. The diagonal line represents no separation because the particle and fluid are proportionally distributed between the upstream and the straight branch of the two downstream capillaries. If there is a positive deviation, more particles are found in the straight capillary or *vice versa*. Therefore the degree of deviation from the diagonal line is a quantitative measure of separation. In Figure 5(a), the number of particles that go to the straight branch is almost proportional to the fractional flow rate to the branch.

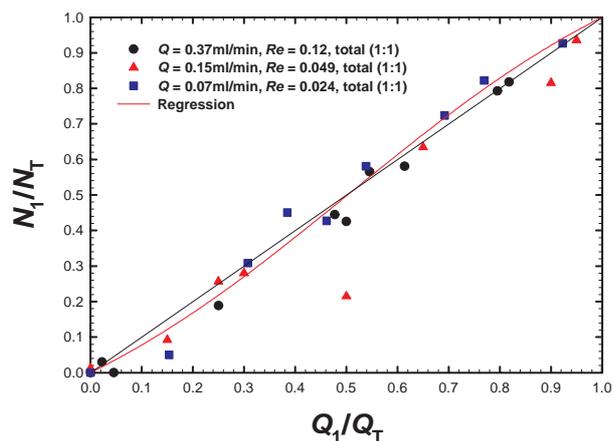
Also in Figure 5(b), the number of large particles that go to the straight branch is also proportional to



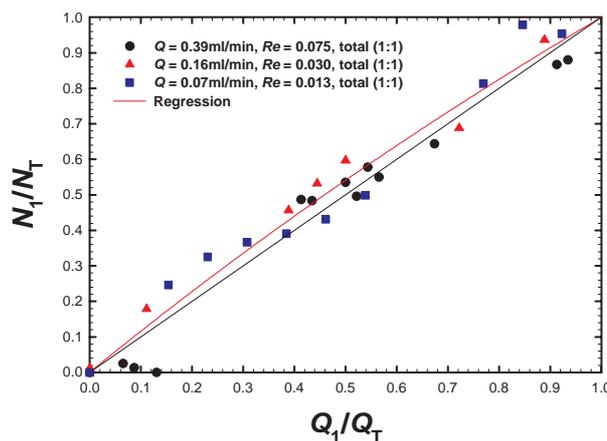
**Fig. 4** T-shaped capillary tube device and the naming of the variables

the fractional flow rate of the particles going to the branch. This means that separation is not possible in this case. Roberts and Olbricht (2003) reported that more particles tend to move to the straight branch when the fractional flow to the straight branch is larger than 0.5, or *vice versa* when the particle size is comparable to the size of capillary. Even though there is a slight tendency of moving more to the straight capillary when the flow to the straight branch is larger than 0.5 as was also observed by Roberts and Olbricht (2003), the small particles suspended in the Newtonian fluid cannot be separated with this geometry. In **Figure 6**, the experimental results are shown for the separation of particles in a non-shear thinning elastic fluid. In this case the curve is not symmetric with respect to the diagonal line meaning that more particles tend to move toward the straight capillary even when the  $Q_1/Q_T$  is less than 0.5. Also the larger particles tend to move to the straight capillary. But the degree of separation is still far from being effective.

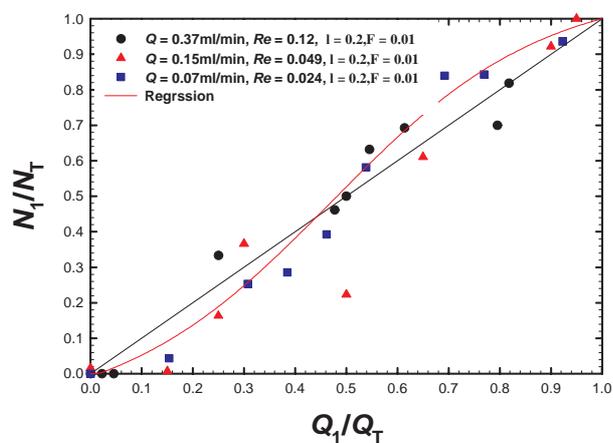
Next we performed experiments with a shear thinning solution without large elasticity. In **Figure 7**, the experimental results are shown for the case of the xanthan gum solution in a water–sugar mixture. In this case the deviation is negative and the deviation from the diagonal line is much larger than the case of non-



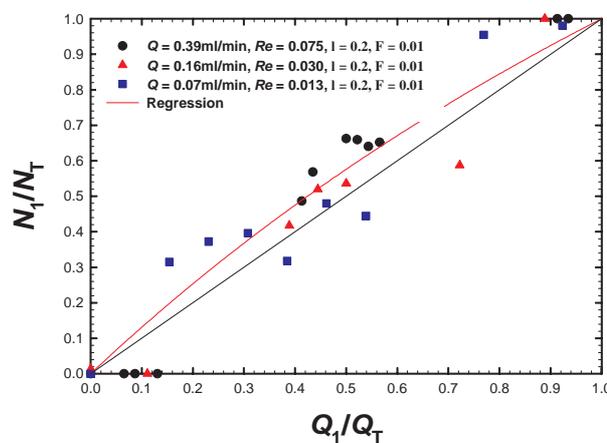
(a)



(a)



(b)



(b)

**Fig. 5** Particle separation in ethylene glycol and glycerin (EG + GL) (52 wt%: 48 wt%): Different symbols represent for the Reynolds number. The volume % of large and small particles are the same (1:1) at 1%. (a) The entire particles; (b) large particles ( $\lambda$  is the ration of the diameter of particle to the diameter of the capillary). The regression has been done to help the reader recognize the trend of separation characteristics. The diagonal line represents no separation. If the data point is located upper part of the diagonal line, more particles are found at the straight capillary and if the data is located at the lower part of the diagonal line, more particles are found in the branched capillary

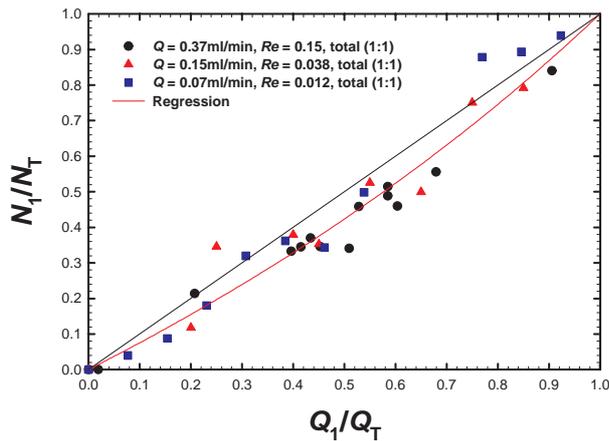
shear thinning elastic solution. In the case of large particles the separation is even stronger. More particles tend to move toward the branched capillary in contrast to the case of non-shear thinning elastic solution. The separation appears to be more effective when the fraction of flow to the straight capillary is smaller than 0.5.

During the experiment we have not observed any aggregation or alignment of particles as observed by

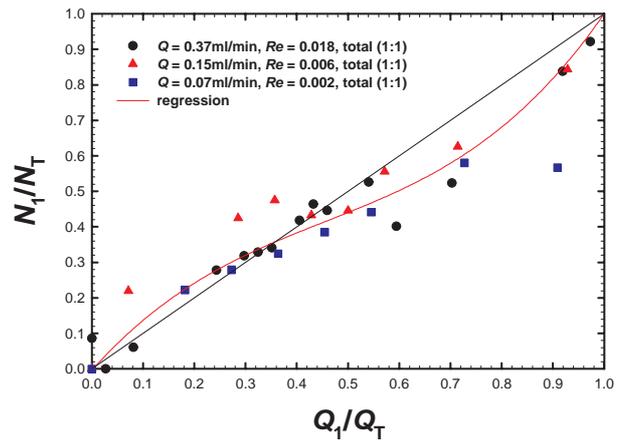
**Fig. 6** Particle separation characteristics in 1000 ppm solution dissolved in EG + GL (52 wt%: 48 wt%): (a) the entire particles; (b) large particles. In this case particles are a little more found in the straight branch

Won and Kim (2004), Jefri and Zahed (1989) and Tehrani (1996). This is because the particle loading is sufficiently small and the entrance length is short enough to get particles aggregated or chained.

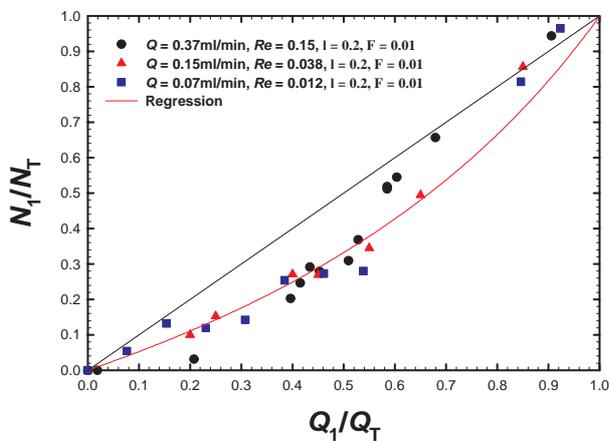
Finally we performed experiments with an elastic and shear thinning solutions. **Figure 8** shows the experimental data for the 2 wt% aqueous solution of polyacrylamide. In this case we did not match the density because the viscosity is sufficiently large hence no sedimentation occurs within the time scale of experimental runs. For the elastic and shear thinning solution, particles are not separated when the flow rate to the straight capillary is less than approximately 0.5 while particles tend to move to the branched capillary when the flow to the straight capillary is larger than 0.5. However, the movement of large particles shows the behavior observed in the shear thinning solution. The



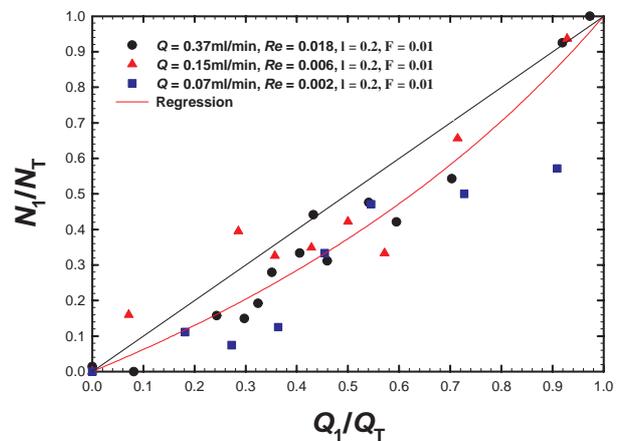
(a)



(a)



(b)



(b)

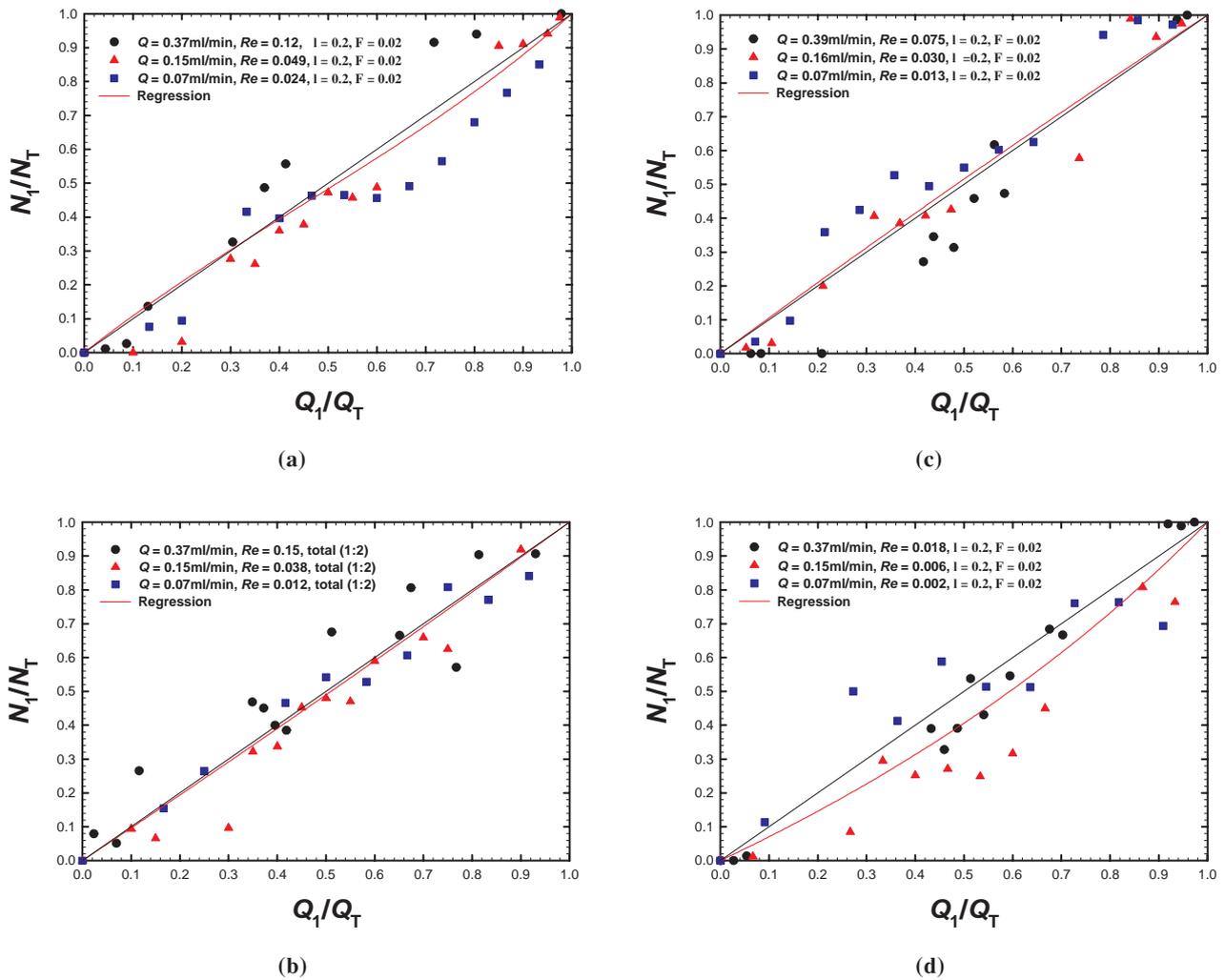
**Fig. 7** Particle separation characteristics in 2000 ppm xanthan gum sugar solution: (a) The entire particles; (b) Large particles. Since the curves are convex downward, more particles are found at the branched capillary

**Fig. 8** Particle separation in aqueous solution of 2% PAAm: (a) the entire particles; (b) large particles. The separation characteristics shows the mixed effect of shear thinning and elasticity

four cases investigated here can be summarized as follows: In the Newtonian fluid, small particles with  $\lambda$  (the ratio of the diameters of particle and capillary) less than 0.2 cannot be separated. In non-Newtonian fluids, particles tend to move in different ways. The elasticity of the fluid makes the particles tend to move more to the straight capillary. By the shear thinning characteristics of the fluid, particles tend to move to the branched capillary. In a viscoelastic and shear-thinning solution, a combined effect is observed.

Next we consider the mechanism of separation. Roberts and Olbricht (2003) considered the partitioning of particles in branched capillaries and showed experimentally that the partitioning characteristics of particles differ from the partitioning of the fluid flow, which led the separation. They also reasoned that their method will be valid only for very dilute suspension

because the particle–particle interaction affects the distribution of particles even at a very low value of particle loading. However, their experimental data were obtained only for a large value of particle-to-diameter ratio in Newtonian fluids. Also their case is limited only to the case of a single kind of particle hence their problem is basically the separation of the solid from the liquid and therefore it is similar to the filtration, while our problem is the separation of small or large particles in a mixture of particles with different sizes in a fluid. Therefore even though the geometry is the same, the mechanism of separation should be fundamentally different. In their case, they assumed that there should be no migration of particles. But in our case, without migration, no separation occurs as in the Newtonian fluids. Therefore we need to consider the migration effect. Before considering the migration



**Fig. 9** Large particle separation characteristics in Newtonian and polymer solution: when the volume fraction of the smaller particles is 0.01 and the volume fraction of the large ones is 0.02: (a) ethylene glycol (EG) + Glycerin (GL) solution; (b) 1000 ppm PAAm (polyacrylamide) solution EG + GL; (c) 2000 ppm xanthan gum + sugar solution; (d) aqueous solution of 2% PAAm

problem in detail, one may consider other forces such as surface related forces and/or Brownian forces that can affect the movement of particles. But since the particle we treat here has such a large size, the surface force related terms and Brownian forces are not expected to contribute to the motion of particles. For example, when the present experimental conditions are employed, the ratio of electrical force and hydrodynamic force is approximately given as (Russel *et al.*, 1989)

$$\frac{\varepsilon\varepsilon_0\zeta^2}{\eta Ua^2} \approx 3 \times 10^{-3} \quad (3)$$

where we assumed that the zeta potential has an order of 10 mV, and the ratio of Brownian force and hydrodynamic force is approximately given as (Russel *et al.*, 1989)

$$\frac{kT}{\eta Ua^2} \approx 4 \times 10^{-10} \quad (4)$$

Therefore we can confirm that the hydrodynamic force is far dominant compared with other forces.

If it is a particle-migration problem due to the particle-particle interaction, the migration length scale that is basically the radius of capillary,  $d$ , is given as follows (Phillips *et al.*, 1992):

$$d = \sqrt{Dt} \sim \sqrt{\dot{\gamma}a^2 \times L/v_{\text{avg}}} \quad (5)$$

where  $D$  is the hydrodynamic diffusivity which scales as  $\dot{\gamma}a^2$  (Leighton and Arivos, 1987),  $t$  is the time span for diffusion,  $\dot{\gamma}$  is the shear rate and  $v_{\text{avg}}$  is the average velocity. Therefore the length,  $L$ , required to separate particles scales as  $L \sim d^3/8a^2$ , where we used the

relationship  $\dot{\gamma} = 8v_{avg}/d$  for the average shear rate. If this scaling is applied, the length that is required to separate particles increases formidably when the particle size becomes small or the diameter of the capillary becomes large due to their high powers. Since the separation was not achieved with a short capillary for the Newtonian fluids, the migration mechanism due to the particle–particle interaction cannot be applied for our problem. The inertial migration mechanism can also be excluded because it also requires a long capillary as is already stated. Therefore the partitioning mechanism should be applied to our problem as well. However, in this case, we additionally include the migration of a single particle across the streamline in a tube flow.

As has been already stated in Introduction of this paper, the migration of a single particle in the Newtonian or non-Newtonian fluid has been well discussed in the literature. When the particle Reynolds number is small, a single particle will not move across the streamline in a tube flow of Newtonian fluid. In the case of a particle in a shear thinning fluid, however, it moves toward the tube wall (Karnis and Mason, 1966; Huang and Joseph, 2000). In a viscoelastic fluid it was reported that a single particle moves toward the tube center (Cox and Mason, 1971; Gauthier *et al.*, 1971). In an Oldroyd-B fluid, which is a non-shear thinning viscoelastic fluid, a single particle tends to move either toward the wall or toward the center depending on the particle–tube diameter ratio. However in the case of suspensions, particles tend to concentrate at the center of tube even in the case of very dilute suspension in the non-Newtonian fluid (Kim, 2001; Tehrani, 1996). If we combine the migration of a single particle with the partitioning mechanism, we can explain how large particles are separated from a mixture of particles with different sizes.

In the case of the Newtonian fluid, particles do not migrate across the streamline. Even though there is a partitioning in favor of the separation, particles in an initially well mixed suspension will keep their radial positions, hence there is no separation. Since particle loading is finite even though it is small, they may be subject to particle–particle interactions which may result in a slight tendency of separation of large particles. To confirm this reasoning we may require a separate research on the particle–particle interaction and subsequent migration of large particles in favor of separation. However it is such a formidable task and is beyond the scope of this paper.

In a shear thinning liquid (xanthan gum 2000 ppm solution), particles tend to move toward the wall and the migration will be faster for large particles. The fast migration was confirmed by the experiment: The larger particles were found near the wall. Then by the partitioning of the flow the large particle tends to move to the branched capillary, which was observed in the ex-

perimental runs. In the case of non-shear thinning viscoelastic fluid (1000 ppm solutions of PAAm), the particles tend to move toward the center which does not contribute to the separation. In a shear thinning viscoelastic solution, the two mechanisms are competing with each other and therefore the separation is not effective. To investigate the effect of the particle–particle interaction, we increased the volume fraction of larger particles to 2% from 1%. **Figure 9** shows that if the particle loading increases, the experimental data do not deviate from the diagonal line, therefore the separation is impossible. This result confirms that the separation is not a result of the particle–particle interaction and the particle–particle interaction even hinders the separation.

In Figures 5 to 9, one can recognize that the data are widely scattered around the fitted line except the case of shear thinning solution. Basically the data are scattered due to the statistical nature of the system. Without any migration mechanism across the streamline in Newtonian and non-shear thinning solutions, the selection of either of the branches is solely determined by the position of the particle at the inlet, and the position of the particle at the inlet is randomly distributed. Since we have shown the raw data, the data appear widely scattered. To remedy this problem, one should count more numbers of particles to average out the scattering. In the case of the shear thinning solution (Figure 7), however, the scattering is not so severe because there is a migration mechanism which makes the problem more deterministic. The different degree of scattering for different fluid also confirms the validity of the combined mechanism of partitioning and migration.

Since it is evident that the separation is caused by the partitioning of the flow together with the migration of non-interacting particles in the flow of shear thinning fluid and is not related to the migration of particles due to the particle–particle interaction, the scaling principle is readily applied to a smaller system. Therefore when we scale it down, the dimensionless velocity profiles are the same and hence the particle separation characteristics will be the same and therefore process miniaturization and intensification are possible.

## Conclusions

In this experimental study, we have investigated the separation of particles with different sizes dispersed in liquids using branched capillaries. We investigated four different kinds of fluids. These were Newtonian fluid, shear thinning fluid, non-shear thinning elastic fluid, and shear thinning and elastic fluid. Only the shear thinning characteristics of the base fluid made the particles tend to be separated and the elasticity prohibited the separation. The mechanism of separation

was deduced to the fast migration of large particles to the wall in the shear thinning fluid flow and the proper partitioning of the flow at the branch. Since the migration of non-interacting particles is fast and the partitioning characteristics are only dependent on the geometry of the channel, we can scale down the system to miniaturize it to achieve the process intensification. In this case we may have to meet some similarity requirements.

#### Acknowledgments

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#### Nomenclature

$a$	= radius of sphere	[m]
$D$	= hydrodynamic diffusivity	[m <sup>2</sup> ·s <sup>-1</sup> ]
$d$	= radius of a capillary	[m]
$G', G''$	= storage and loss modulus	[Pa]
$K$	= Boltzmann constant	[J·K <sup>-1</sup> ]
$L$	= entrance length of capillary	[m]
$N$	= number of particles	[—]
$Q$	= flow rate	[m <sup>3</sup> ·s <sup>-1</sup> ]
$T$	= temperature	[T]
$U$	= representative velocity	[m·s <sup>-1</sup> ]
$v_{\text{avg}}$	= average velocity	[m·s <sup>-1</sup> ]
$\epsilon$	= dielectric constant	[—]
$\epsilon_0$	= permittivity of vacuum	[F·m <sup>-1</sup> ]
$\dot{\gamma}$	= shear rate	[s <sup>-1</sup> ]
$\eta$	= viscosity	[Pa·s]
$\zeta$	= zeta potential	[V]

#### <Subscript>

1, 2	= branch number
T	= total

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