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Drag-reduction effectiveness of xanthan gum in a rotating disk apparatus

J.-I. Sohn[a], C.A. Kim[b], H.J. Choi[b,*], M.S. Jhon[c]

[a]*Department of Chemistry, Hallym University, Chunchon 200-702, South Korea*
[b]*Department of Polymer Science and Engineering, Inha University, Inchon 402-751, South Korea*
[c]*Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA*

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Abstract

Drag reduction, achieved by introducing a minute amount of a homologous series of polysaccharide xanthan gum in an aqueous solution, is investigated using a rotating disk apparatus. In this paper, the ultrasonic degradation method is adopted to obtain different fractions of molecular weights of xanthan gum. The dependence of drag reduction on various factors, including polymer molecular weight, polymer concentration, rotational disk speed, ionic strength of solution, and temperature, is investigated. Drag reduction induced by the xanthan gum is found to increase with polymer concentration, reaching a maximum at its critical concentration, while the concentration required for maximum drag reduction decreases with increasing molecular weight. Polymer degradation due to the high shear forces, which occur during drag-reduction experimentation, is also found to increase with turbulence intensity. Furthermore, tests show that xanthan gum is a suitable drag reducer even at relatively high temperatures.

Keywords: Xanthan gum; Drag reduction; Polymer degradation; Turbulent flow; Polysaccharide

1. Introduction

The addition of a very small amount of high-molecular-weight polymer into a liquid can cause a drastic reduction in the frictional drag in turbulent flow. This turbulent drag-reduction phenomenon implies that polymer solutions undergoing flow in a pipe would require a lower pressure drop to maintain the same flow rate and that a higher flow rate would be obtained for the same pressure gradient if such an additive were applied. Therefore, this phenomenon provides considerable motivation for diverse research in order to discover its mechanism and applications, including the transport of crude oil (Burger, Chron & Perkins, 1980), increasing volumetric flow rate of water in fire fighting (Fabula, 1971), increasing the speed of boats and torpedoes (Fabula, Green & Madison, 1980; Hoyt, 1972), enhancing water supply and irrigation systems (Sellin, Hoyt & Scrivener, 1982; Singh, Singh, Deshmukh, Kumar & Kumar, 1989), the hydraulic transportation of solid particle suspensions (Golda, 1986), improving cooling and heating circulation systems (Myska & Zakin, 1997), inhibiting fuel misting in aircrafts (Hoyt, 1980), and enhancing ocean thermal energy conversion systems (Kim, Sung, Choi, Kim, Chun & Jhon, 1999).

Even though its mechanism is not yet entirely understood (Armstrong & Jhon, 1983, 1984), it is generally accepted that drag reduction is associated with the viscoelasticity of polymer solutions. De Gennes (1990) introduced an elastic theory for drag reduction to discuss the properties of homogeneous, isotropic, three-dimensional turbulence in the presence of polymer additives. The central idea of De Gennes' "cascade theory," limited to linear flexible chains in a good solvent, is that polymer effects on small scales are not described via the shear viscosity, but instead by the elastic modulus. The importance of elastic properties in the description of the drag-reduction mechanism was also examined by Armstrong and Jhon (1984). Adopting a simple model to study the coupling between turbulence and dissolved polymer molecules, they related molecular dissipation to friction factors by a self-consistent theoretical approach.

Among various additives such as polymers, surfactant solutions, and fiber particles, polymers are considered the most effective drag reducing agent. High-molecular-weight poly(ethylene oxide) (PEO) and polyisobutylene have been widely used as drag reducers (Berman, 1978; Kim, Lee, Choi, Kim, Kim & Jhon, 1997; Sellin et al., 1982; Usui, 1990). However, the use of these polymers in practical applications has been severely limited due to their poor mechanical stability in turbulent flow. The search for shear-stable drag-reducing polymers continues to be a challenging task.

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[*] Corresponding author. Tel.: +82-32-860-7486; fax: +82-32-865-5178.
E-mail address: hjchoi@inha.ac.kr (H.J. Choi).

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Certain industrial polysaccharides (Chakrabarti, Seidl, Vorwerk & Brunn, 1991a,b; Deshmukh & Singh, 1987), such as hydroxypropylguar, guar gum, and xanthan gum, have been found to be reasonably shear-stable drag-reducing agents. The advantage of polysaccharide polymers is their high mechanical stability against degradation when compared to flexible synthetic polymers with similar molecular weights; however, they are highly susceptible to biological degradation. To overcome this disadvantage, graft copolymerization methods have been introduced (Deshmukh & Singh, 1986, 1987; Deshmukh, Chatuvedi & Singh, 1985). Graft copolymers using guar gum enhance both drag-reduction effectiveness and shear stability.

Xanthan gum, an extracellular polysaccharide produced by the bacterium *Xanthomonas campestris*, consists of 1,4-linked β-D-glucose residues, having a trisaccharide side chain attached to alternate D-glucosyl residues (Katzbauer, 1998). The backbone of the polymer is similar to that of cellulose. It has been widely used in different applications, e.g. as a viscosity-enhancing agent in foods, in the cosmetics and pharmaceutical areas, and in oil drilling fluids and enhanced oil recovery. It even has the potential for use in coal water mixtures (Katzbauer, 1998; Podolsak, Tiu, Saeki & Usui, 1996).

The aim of this study is to characterize xanthan gum as a drag reducer and to investigate the effects of molecular parameters on drag reduction using a rotating disk apparatus (RDA). Most of the previous drag-reduction measurements have been performed using a pipe flow apparatus, where turbulence is generally produced by driving the fluid through a looping channel. In contrast, with an RDA, turbulence is produced by the motion of a surface located within a fluid, i.e. the rotating disk system is used to describe external flow that includes flow between flat plates as well as flow around submerged objects. The RDA allows convenient measurement of the long-term drag-reduction properties of polymer solutions.

2. Experimental

2.1. Materials and solution preparation

All measurements described in this paper were performed using xanthan gum purchased from Sigma Chemical Co. (St. Louis, MO, USA). The molecular weight of this material has been reported to be a few million for a single-stranded material (Sato, Norisuye & Fujita, 1984).

Because only a single molecular weight of xanthan gum is available commercially, the ultrasonication method was adopted to produce several different molecular weights of xanthan gum (Kim, Choi, Kim & Jhon, 1998) by polymer chain scission. Ultrasonication has been adopted as the best means to obtain several fractions without any structural or chemical modification (Chun & Park, 1994). Sample solutions were exposed to 750 W of 28 kHz sound with a custom-made ultrasonicator for a specified time according to the degree of chain scission desired. Stock solutions of 0.5 wt% aqueous xanthan gum solutions were prepared in a flask by mildly stirring with a magnetic bar for a week without any bactericides or other background electrolytes. These prepared solutions were then stabilized for one day before use. Our ultrasonicator is equipped with a controllable water bath. The flask with the xanthan gum solution was fixed in the bath, and the ultrasound was applied for specific times (15, 30 and 60 min at constant temperature). We then obtained the viscosity-average molecular weight of the fractions by measuring the intrinsic viscosity $[\eta]$ in deionized water using an Ubbelohde capillary viscometer (Chun & Park, 1994) and applying the Mark–Houwink equation reported by Holzwarth (1978)

$$[\eta] = \lim_{C\to 0} \frac{\eta_{\mathrm{sp}}}{C} = 6.6\times 10^{-6}(\bar{M}_{\mathrm{V}})^{1.35}\ [\mathrm{ml/g}] \tag{1}$$

The specific viscosity η_{sp} was obtained from the relative viscosity which is the ratio of the viscosity of xanthan gum solution at a certain concentration (C) to that of the solvent. Using the Ubbelohde viscometer (Schott-Geräte, Germany, Capillary No. 52610/I), the flow times of each xanthan gum solution and pure solvent (deionized water) were measured at several different polymer concentrations. The temperature was accurately kept at 20.00 ± 0.005°C by a constant temperature circulating system (Haake F6, Germany). The plot of η_{sp}/C versus C gave a straight line, the intercept of which was $[\eta]$. Furthermore, to observe the salt effect on drag reduction, an additional stock solution with NaCl was prepared for unsonificated xanthan gum.

2.2. Observation of drag-reduction effects

Drag-reduction measurements were performed with the same RDA used by Kim et al. (1997, 1998). The RDA consists of a stainless steel disk whose dimensions are 10.1 cm in diameter and × 0.32 cm in thickness, enclosed in a cylindrical thermostatically controlled container, which is made of stainless steel and whose dimensions are 16.3 cm in inner diameter × 5.5 cm in height. The volume of solution required to fill the entire container is approximately 1020 cm^3. The rotational velocity of the disk was controlled by a speed controller (Cole Parmer Master Servodyne Unit), and the torque was measured by a multimeter and then stored on a PC via an A/D converter. The temperature of the system was maintained at 25 ± 0.5°C by a constant temperature circulating apparatus, and the rotational velocity of the disk was measured by a digital tachometer.

The required amount of stock solution was measured and then carefully injected directly into the turbulent flow. A flow visualization experiment to determine whether the fluid was uniformly mixed after the addition of the stock solution was performed with the apparatus used by Yang, Choi, Kim, Kim and Jhon (1991). We observed complete mixing within several seconds and obtained reproducible

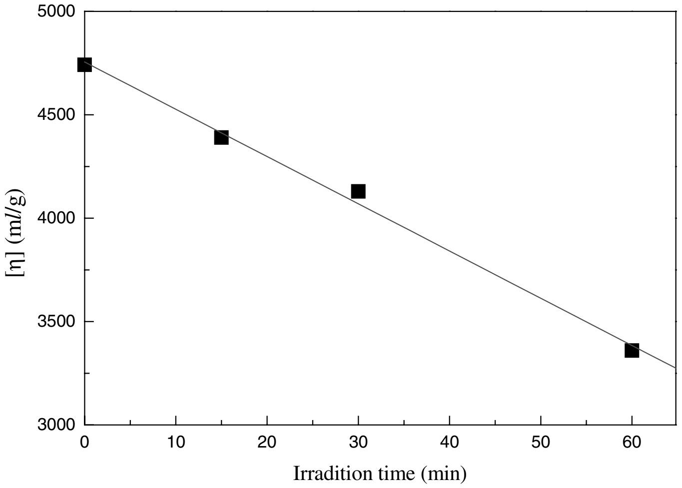

Fig. 1. $[\eta]$ of xanthan gum as a function of ultrasonic irradiation time at 20°C.

results for each run. However, experimental error could have occurred before uniform mixing was completely achieved.

Drag reduction is known to be related only to frictional drag, while the RDA measures the total drag reduction. Note that one typically studies frictional drag for an internal flow, whereas external flows measure total drag (friction plus form drag). Because of this difference, a maximum of 80% of the drag reduction can be obtained for pipe flow, whereas a rotating disk flow generally produces approximately 50% of the maximum drag reduction.

The disk rotated at a fixed rotation speed, and the resultant torque reduction was measured as a function of time. The percent drag reduction (%DR) to quantify the drag-reduction efficiency was calculated via the torque on the disk in the solvent and that in the polymer solution by (Kim, Kim, Lee, Choi & Jhon, 2000)

$$\%\mathrm{DR} = \frac{T_\mathrm{S} - T_\mathrm{P}}{T_\mathrm{S}} \times 100 \qquad (2)$$

where T_P is defined as the millivolt output (torque) for the polymer solution, and T_S is defined as the torque for the solvent.

Flow characteristics in this RDA can be described by the rotational Reynolds number (N_Re)

$$N_\mathrm{Re} = \frac{\rho R^2 \omega}{\mu} \qquad (3)$$

where ρ is the fluid density, μ the fluid viscosity, R the radius of the disk, and ω the angular velocity of the disk. The critical N_Re is 3×10^5, which correspond to a rotation speed of 1050 rpm (for a water system) in our RDA.

All of the experiments in this study were performed at a fixed rotation speed of 1800 rpm and temperature of 25°C, unless otherwise specified.

3. Results and discussion

Xanthan gum is one of the most important industrial biodegradable polyelectrolytes, and its conformation is known to be dependent on both temperature and ionic strength. At high temperature and low ionic strength xanthan exists in solution as a disordered coil. Upon cooling and/or the addition of salt, it becomes ordered. There is a general belief that this ordered structure involves two-chains (Sato et al., 1984), each of five-fold helical symmetry, although it is still not clear whether they are packed together co-axially or side-by-side. As has been found for other polyelectrolytes that convert from a disordered coil state at high temperature to an ordered form at low temperature, the transition-midpoint temperature (T_m) increases systematically with increasing ionic strength (I). Quantitatively, there is a linear relationship between $1/T_\mathrm{m}$and $\ln I$. Qualitatively, increasing the solution ionic strength promotes an ordered structure by screening intramolecular and intermolecular repulsion. In the case of xanthan gum, the electrostatic repulsions are between the charged groups on the individual side-chains, which must pack together along the polymer backbone to allow the helical structure to form, and between the two strands of the duplex. Therefore, the viscosity of xanthan gum is function of temperature, salt type, and concentration (Capron, Brigand & Muller, 1997; Milas & Rinaudo, 1979; Norton, Goodall, Frangou, Morris & Rees, 1984). Furthermore, because the molecular characterization becomes greatly complicated by the presence of cations and a microgel fraction (Arendt & Kulicke, 1998), it is difficult to obtain xanthan gum samples with different molecular weights. Fig. 1 illustrates the intrinsic viscosities of four different fractions as a function of the ultrasonic irradiation time, where data at the initial time indicate that of the unsonicated xanthan gum. $[\eta]$ is a function of the polymer molecular weight as given in Eq. (1). Therefore, the viscosity-average molecular weight is

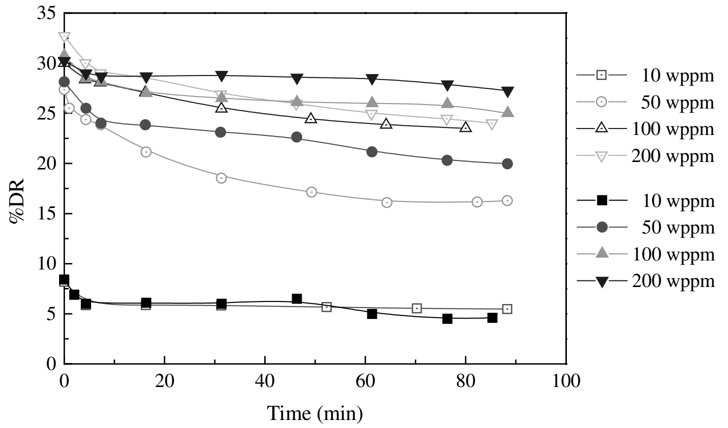

Fig. 2. Influence of concentration on %DR as a function of time of unisonicated xanthan gum in deionized water (open symbols) and NaCl (500 wppm) solution (filled symbols).

obtained from $[\eta]$. $[\eta]$ decreases with ultrasonic irradiation time as shown in Fig. 1. The following $\bar{M}_V$ were obtained: 3.61×10^6 for unsonicated xanthan gum, 3.41×10^6 after 15 min irradiation, 3.25×10^6 after 30 min irradiation, and 2.80×10^6 after 60 min irradiation, and we index them XGU, XG15, XG30 and XG60, respectively. From this experimental result, the ultrasonication irradiation is confirmed to be an excellent method for obtaining different molecular weight fractions.

From drag-reduction measurements for various molecular weight fractions, we also determined the stability (or mechanical degradation) of xanthan gum chains in turbulent flow. Furthermore, to study salt effects, we performed drag-reduction experiments using an NaCl solution (500 wppm) as a solvent. Fig. 2 shows the drag reduction obtained for different concentrations of xanthan gum as a function of time both in deionized water and salt solution. As indicated in this figure, xanthan gum behaves as a more shear-stable drag-reduction agent in the deionized water, as well as in the salt solution, than most flexible synthetic polymers (Deshmukh et al., 1985; Kim et al., 1998). At a relatively low concentration, such as 10 wppm, no difference in %DR is observed. However, at higher concentrations, especially above 50 wppm, the drag-reduction behavior exhibits greater stability in the salt solution than in deionzed water. The ordered conformation of xanthan gum, which is responsible for the extraordinary stability of the polymer, is enhanced by salt. Note that drag reduction is known to be a function of molecular weight, chain linearity and flexibility, and molecular expansion in solution (Kim, Long & Brown, 1986). In addition, Kim et al. (1986) reported unusual drag-reduction characteristics for polyacrylic acid in the presence of NaCl. They explained this unusual behavior by the conformation transition to form molecular aggregates due to interchain bonding forces (e.g. hydrogen bonding). By adopting this argument, we suggest that the addition of NaCl induces not only conformational changes involving hydrogen bonding, but also the drag-reduction recovery of xanthan gum.

We also examined the effect of temperature on drag

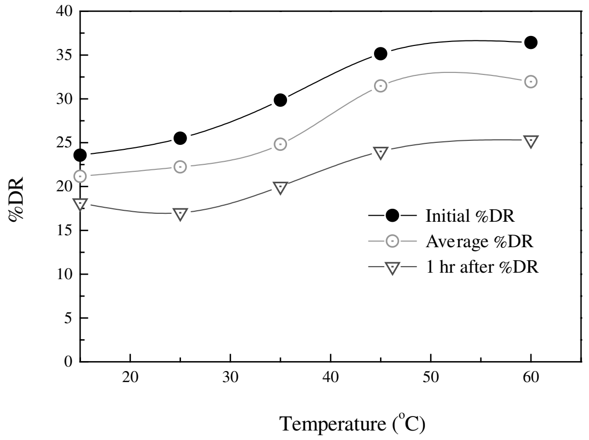

Fig. 3. Temperature dependence of %DR with 50 wppm unsonicated xanthan gum solution. "Initial %DR" and "Average %DR" indicate the data obtained immediately after the polymer was injected into the turbulent flow and data averaged from the initial %DR and 1 h after %DR, respectively.

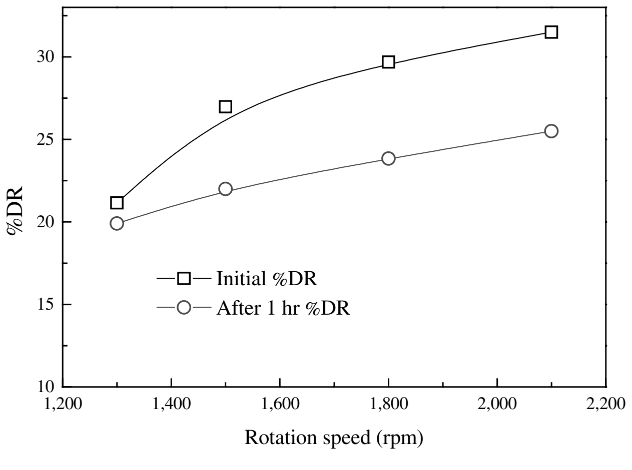

Fig. 4. %DR of unsonicated xanthan gum versus rotation speed (turbulent intensity) at 100 wppm concentration.

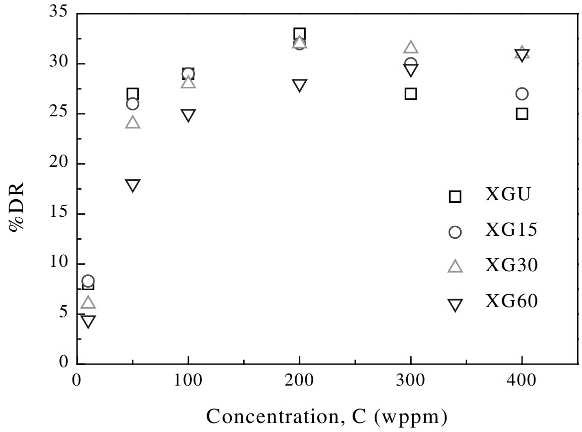

Fig. 5. Initial %DR versus xanthan gum concentration at four different molecular weights at 1800 rpm.

reduction for xanthan gum as shown in Fig. 3. The sigmoidal increase in drag reduction with increasing temperature over the approximate range 30–50°C can be attributed to the thermally-induced order–disorder transition of xanthan gum, with conversion of aggregated helices to a larger number of individual coils each stiffened and expanded by intramolecular electrostatic repulsion (Smidsrød & Haug, 1971). A similar increase with increasing temperature has been observed (e.g. Arendt & Kulicke, 1998; Chen & Sheoppard, 1980) in the viscosity of xanthan gum solutions at high shear rates. The ordered form is, however, less susceptible to degradation; Chen and Sheoppard (1980) found that, when exposed to high shear (5000 s^{-1}), xanthan gum showed less degradation in salt solution than in distilled water. Based on these results, we can postulate that the drag reduction of xanthan gum has a higher value in the range 50–60°C than that at lower temperatures.

Fig. 4 shows both the initial %DR and %DR after 1 h as a function of rotation speed for the unsonicated xanthan gum solution at 100 wppm concentration. The difference between the initial drag reduction and the 1 h drag reduction gradually increases with rotation speed. That is, the drag reduction increases with the rotation speed, and also the degradation of molecules is accelerated with increased speed. This is consistent with the fact that the drag-reduction effect is enhanced with increased turbulence intensity.

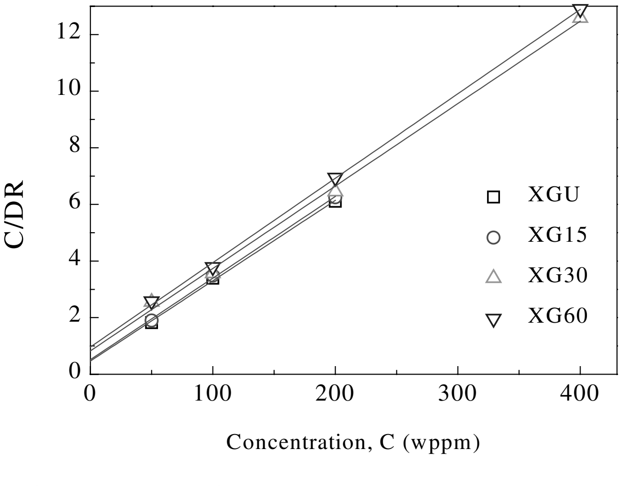

Fig. 6. *C*/DR versus *C* for four different molecular weights of xanthan gum at 1800 rpm.

Because the drag reduction results from the sum of contributions from individual polymer molecules, we measured the drag reduction as a function of concentration. Fig. 5 shows the dependence of percent drag reduction on four different molecular weights of xanthan gum as a function of polymer concentration up to 400 wppm. Maximum drag-reduction values for different molecular weights of xanthan gum are: 31% for $\bar{M}_V = 2.80 \times 10^6$ (XG60), 32.1% for $\bar{M}_V = 3.25 \times 10^6$ (XG30), 32.3% for $\bar{M}_V = 3.41 \times 10^6$ (XG15), and 33.0% for $\bar{M}_V = 3.61 \times 10^6$ (XGU). This clearly indicates that the concentration required for maximum drag reduction decreases with increasing molecular weight.

To find an empirical relationship between the drag-reduction efficiency and polymer solution properties for both water- and oil-soluble polymers, Choi and Jhon (1996) introduced a three-parameter empirical relationship between drag reduction (DR) and concentration (*C*) to provide a universal correlation. This relationship, which accounts for the concentration dependence of drag reduction, has the following form (Choi, Kim & Jhon, 1999; Kim et al., 1998) at a fixed N_{Re}:

$$\frac{C}{\mathrm{DR}} = \frac{K[C]}{\mathrm{DR}_{\max}} + \frac{C}{\mathrm{DR}_{\max}} \tag{4}$$

where $\mathrm{DR}_{\max}$ is the maximum drag reduction, *K* is a characteristic parameter which depends on the polymer–solvent system, and [*C*] is the intrinsic concentration (in wppm) defined by

$$[C] = \frac{\mathrm{DR}_{\max}}{\lim_{C \to 0} (\mathrm{DR}/C)} \tag{5}$$

The quantity in the denominator is the intrinsic drag reduction, which is a measure of the drag reduction per unit concentration at infinite dilution. Moreover, [*C*] is inversely related to the molecular weight of the polymer (Yang, Choi, Kim, Kim & Jhon, 1994). Eq. (4) shows that there is a linear relationship between *C*/DR and *C* up to the optimum concentrations of each molecular weight, and this relationship is valid for most drag reducing polymers for any flow geometry (pipe flow or RDA). The linear correlation between polymer *C* and *C*/DR for four different molecular weights of xanthan gum in a range of conditions close to the maximum drag reduction is illustrated in Fig. 6. $\mathrm{DR}_{\max}$ can be obtained from the reciprocal of the slope, and the intercept value yields [*C*] when multiplied by the quantity $\mathrm{DR}_{\max}/K$. We obtained the *K* value to be 1.2 from Virk's universal curve for drag reduction (Kim et al., 1998). Hunston and Zakin (1980) found that for various

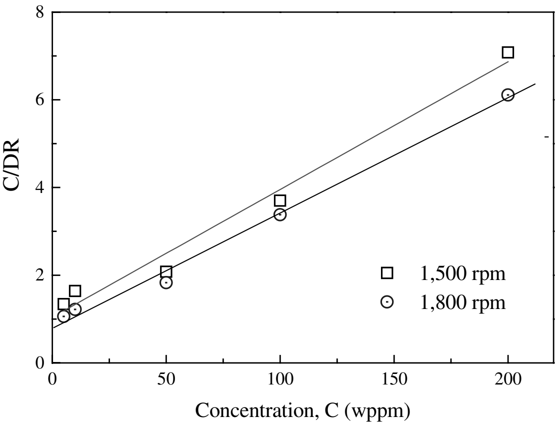

Fig. 7. *C*/DR versus *C* for unsonicated xanthan gum at two different rotation speeds.

polymer–solvent systems, more efficient materials have a larger DR_{max} and a smaller [*C*]. As shown in Fig. 6, the intercept values decrease with molecular weight which confirms that good drag reducers exhibit low values of intrinsic concentration. From the measurement of the effect of molecular weight on the intrinsic concentration, we obtained the intrinsic concentration: 13.8 wppm XGU, 15.0 wppm for XG15, 23.8 wppm for XG30 and 27.1 wppm for XG60. Furthermore, we determined the effect of turbulence intensity on the intrinsic concentration as shown in Fig. 7. As previously discussed, the drag-reduction effect increases with the turbulence intensity.

From the results of McCormick and his collaborators (McCormick, Hester, Morgan & Safieddine, 1990; Mumick, Welch, Hester & McCormick, 1992; Mumick, Welch, Salazar & McCormick, 1994), various model polymers were found to collapse to a universal curve for drag reduction when normalized by the hydrodynamic volume fraction of the polymer solution. They reported that volume fraction normalization allows the comparison of drag-reduction efficiencies of polymers of widely differing structures, compositions, and molecular weights. Fig. 8 is a plot of percent drag reduction per unit volume fraction versus polymer hydrodynamic volume fraction. The polymers that yield the greatest values of (%DR/[η]*C*) at a specific volume fraction ([η]*C*) are the most efficient drag reducers. As expected, the data in Fig. 8 collapse to a single line (a universal curve is obtained) independent of the molecular weight.

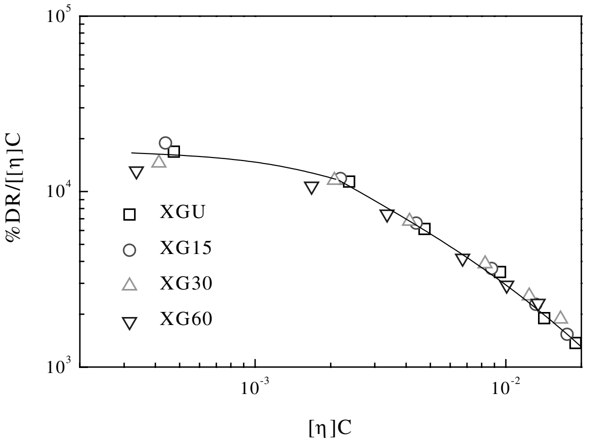

Fig. 8. Drag reduction per unit volume fraction (%DR/[η]*C*) versus volume fraction ([η]*C*) of xanthan gum in deionized water.

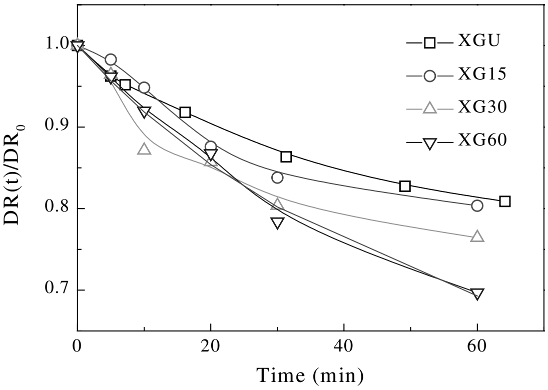

Fig. 9. Relative drag reduction of xanthan gum versus time with 100 wppm solution for four different molecular weights.

As discussed before, xanthan gum exhibits high shear stability. We further examined the resistance of xanthan gum to degradation. Drag reduction has been found to be directly proportional to the relative molecular weight of polymers. These results can be described by the relative drag reduction (DR efficiency ratio)

$$\frac{DR(t)}{DR_0} = \frac{M(t)}{M_0} \tag{6}$$

where DR(*t*) is percent drag reduction at time *t*, *M*(*t*) is the effective number-average molecular mass at time *t*, and the index 0 pertains to the beginning of flow. The degradation profiles for different molecular weight xanthan gum in deionized water are shown in Fig. 9, where the relative drag reductions for different molecular weights retain above 70% of their initial level of drag reduction after 1 h. Under similar conditions, PEO retains below 40% of its initial drag reduction (Kim et al., 1999), and xanthan gum has much better resistance to shear degradation compared to a typical water-soluble drag reducer (i.e. PEO).

4. Conclusions

The drag-reduction behavior of several different molecular weights of xanthan gum obtained from the ultrasonic irradiation method is studied. Drag reduction increases with polymer concentration until a maximum drag reduction is reached, and the concentration required for maximum drag reduction decreases with molecular weight. Drag-reduction efficiency is closely related to molecular parameters, such as stress level, temperature, polymer species, molecular weight, and polymer–solvent interactions. Temperature is an important factor, because the polymer conformation changes with temperature. Xanthan gum molecules in deionized water exhibit higher drag reduction at temperatures in the range 50–60°C than at room temperature, since in this temperature range xanthan gum molecules change from aggregated helices to individual coils expanded by intramolecular electrostatic repulsion.

Higher drag reduction is obtained by increasing rotation speed, however noticeable polymer degradation is observed after some time. Furthermore, different molecular weights of xanthan gum correlate very well with the empirical polymeric drag-reduction equations, such as Virk's linear curve and the hydrodynamic volume correlation. Finally, xanthan gum is confirmed to be a suitable drag-reduction agent in the case of relatively high-temperature and long-term applications.

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