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journal homepage: www.elsevier.com/locate/jiec1 Effects of hydrophobic modification of xanthan gum on its turbulent
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ABSTRACT

The effects of different structures of biopolymeric xanthan gum (XG) on its drag reduction (DR) in turbulent flow in an aqueous solution with pristine XG and XG modified by grafting octylamine were examined. Owing to the more rigid conformation of hydrophobically modified XG, its chemical modification allowed a study of the impact of chain flexibility on DR and mechanical degradation. The measurements were carried out at four XG concentrations using a high-precision rotating disk apparatus as a function of time at 1980 rpm. The results showed that the flexibility and concentration with a similar molecular weight affect the DR. Time-dependent DR efficiency was fitted and rheological characteristics of the XG (0.01 wt%) were examined using a rotational rheometer. The influence of the flexibility of the XGs on turbulent DR were compared and analyzed according to these observations.

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6 Introduction

7 The addition of miniscule amounts of polymers, surfactants,
8 and fiber particles to a liquid can cause a remarkable decrease in
9 the skin frictional drag in turbulent flow [1–5]. Among the various
10 active turbulent drag reducing agents, flexible, linear, and long-
11 chain high molecular weight polymers have been reported to
12 produce an effective drag reduction (DR) output. This DR
13 phenomenon explains that polymeric fluids, even with an
14 extremely dilute polymer concentration in pipe flow commonly
15 require a lower pressure drop to retain the same volumetric flow
16 rate, resulting in a reduced pumping energy [6]. The DR has been
17 the subject of many studies since its first report [7] because of its
18 academic interest and potential engineering applications, includ-
19 ing the pipeline transportation of crude oil [8], firefighting [9],
20 flood water disposal [10], biomedicine [11], transportation of
21 slurries and suspensions [12,13], and cooling and heating systems
22 [14].

23 Although diverse theories and mechanisms between turbulent
24 flow and polymer chains have been reported, the precise
25 mechanism has not been explained comprehensively. Of those,
26 an elastic theory of turbulent DR was suggested to explain the

characteristics of homogeneous and isotropic three-dimensional
turbulence in the presence of polymer additives except for any wall
effects using a cascade theory [15]. According to this theory,
restricted linear-flexible chains play an important role in the
polymeric effects on a small scale that are interpreted by the elastic
modulus not by the shear viscosity. In a description of the DR
mechanism, the significance of the elastic property was analyzed
by choosing a simple model to examine the turbulence and
polymer-solution molecules. In addition, the turbulent DR was
described with the solvation of polymer chains and the formation
of correspondingly stable domains [16], in which the flexibility of
the polymers was found to assist in domain formation. Studies
using poly (ethylene oxide) (PEO) and polyisobutylene with a high
molecular weight as drag reducers in aqueous and organic
medium, respectively, have been conducted [17]. While their
use as turbulent DR agents for substantive applications are limited
by their poor mechanical stability in turbulent flow, industrial
polysaccharides, such as xanthan gum, guar gum, karaya gum, and
hydroxypropyl guar have been found to be shear-stable DR agents
along with various other engineering applications [18]. Although
polysaccharides are greatly sensitive to biological degradation,
they have high mechanical stability and resistance to degradation
compared to flexible polymers with similar molecular weights.

Among the polysaccharide gum family, xanthan gum (XG) is an
extracellular anionic polysaccharide generated by the bacteria
Xanthomonas campestris and composed of 1,4-linked- β -D-glucose

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with a trisaccharide side chain including a D-glucosyl unit, while its backbone is similar to that of cellulose. Furthermore, because XG has carboxylic functionality, it is possible to make the chemically modified XG in a regulated and specific manner. Roy et al. [19] developed a procedure to chemically modify xanthan by grafting octylamine groups onto the carboxylic acid functionality of XG, demonstrating that hydrophobically modified XG has a more rigid conformation in aqueous solutions [20].

Therefore, this study focused on the effects of the flexibility of XG on the turbulent DR efficiency. Most DR experiments have been carried out using a pipe flow apparatus. On the other hand, the present study used a rotating disk apparatus (RDA), where turbulence is generated by the motion of a rotating surface. The RDA is considered to be a convenient device for examining the long-term DR properties of polymer solutions. Furthermore, it can be also noted that in addition to the recent study of effect of salt on DR [21], various kinds of water-soluble polymer based drag reducers were well summarized [22].

Experimental

To analyze turbulent DR, most studies used pipe flow [23–25], which generates a pressure-driven flow in a surrounded channel as an internal flow. In the rotating disk apparatus (RDA) system, DR in an external flow can be measured easily as a function of time. The RDA equipment was comprised of a stainless steel disk, 14.5 cm in diameter and 0.32 cm in thickness, and was enclosed in a cylindrical, thermostatic-controlled container using a circulating fluid bath at 25 °C. A speed controller was used to control the rotational velocity of the disk, while an electric transducer was used to observe the torque on the disk rotating at 1980 rpm, producing a specific Reynolds number, N_{Re} .

In terms of external flow in the RDA, turbulent flow occurs when $N_{Re} > 3 \times 10^5$, corresponding to a disk rotational speed of $\omega > 570 \text{ rpm} \times 2\pi$. Here, N_{Re} is described as

$$N_{Re} = \frac{\rho r^2 \omega}{\mu} \quad (1)$$

where ρ is the fluid density, μ is the fluid viscosity, r is the radius of the disk and ω is the angular velocity [rad/s, $2\pi \times (\text{revolution per minutes})/60$]. The temperature of RDA was fixed to 25 °C using a circulating fluid bath and the rotation speed was set to 1980 rpm. Once the torque required to rotate the disk in a pure solvent (T_s) was measured, the percentage DR (%DR) efficiency was estimated by measuring the corresponding torque required at the same rotation-speed in a solution with the added dilute-polymer (T_p) using the following equation:

$$\%DR = \left[\frac{(T_s - T_p)}{T_s} \right] \times 100 \quad (2)$$

Two types of XG with molecular weight about one million g/mol were adopted for this DR test, in which the pristine XG was provided by Danisco (France), with the degrees of acetate and pyruvate substitution determined by ¹H NMR equal to 0.87 and 0.49, respectively [19]. On the other hand, for the hydrophobically modified XG, it was produced by chemical grafting of a hydrophobic-octylamine chain onto the carboxylic group of the pristine XG via a carbodiimide-mediated peptide coupling reaction in water [19]. By conducting the modification process of XG, the properties of modified XG were changed because of the alkyl chains; the number of hydrogen bonds, rigid-helix conformation, associating properties, and rheological characteristics, depending on the grafting density. More detailed information on modified XG can be found elsewhere [19]. In particular, the sample used in this experiment was X³⁰C₈, where C₈ means that octylamine

(8 carbons) was grafted onto the xanthan backbone and 30 corresponds to the percentage grafting density with respect to the repeating unit of XG.

Both XG and modified XG solutions with several different concentrations (10, 50, 100 and 200 ppm) were prepared by mildly stirring for 2 days in deionized water. The prepared solutions were then stabilized for 1 day before starting the DR measurement. The %DR was obtained as a function of time by placing the polymer solutions directly into the turbulent flow caused by the RDA.

Furthermore, to determine how its modification affects the rheological characteristics of each polymer solution with 0.01 wt. % at 25 °C, a rotational rheometer (Anton-Paar, MCR300) with a Couette cell geometry (DG 26.7) was adopted. The steady shear viscosity measurements were conducted as a function of the shear rate in the range, 10^{-3} – 10^2 s^{-1} .

Results and discussion

The rheological characterization of the two XGs was influenced by the ordered molecular conformation [26]. Indeed, by grafting a hydrophobic group onto the backbone of XG, the modified XG chain became more rigid than the native XG. Note that an increase in shear viscosity would become more significant for a rigid polymer than for a flexible one [27], because more rigid-particles show greater resistance to deformation. This was confirmed in Fig. 1 in that modified XG showed a larger increase in shear viscosity (approximately 10 times more viscous within the linear regime) and shear stress as function of the shear rate. For both XG solutions, shear-thinning behavior was clearly observed over a wide range of shear rates. The decrease in shear viscosity with increasing shear rate was attributed to the alignment of polymeric molecules within the flow field [28]. In the case of salt effect on shear viscosity of the XG solution, the decrease of shear viscosity was also observed with the increase of salt concentration due to the increased compact helical backbone conformation, resulting in a decreased hydrodynamic volume and a lower viscosity [21].

To analyze the effects of rigidity on the DR efficacy with two types of XG, the %DR was measured at four different concentrations (10, 50, 100, and 200 ppm) as a function of time in deionized water with a rotational speed of 1980 rpm at 25 °C. As shown in Fig. 2, the initial %DR, which was measured immediately after the stock XG solution was injected into the RDA, increased with increasing XG concentration due to the increase in the number of accessible polymeric drag reducers. Over the entire region, the %DR of native XG had a higher value than that of modified XG. This could be related to the flexibility of the polymer chains because a more rigid polymer leads to less sensitivity to the high shear-condition with less elasticity of its backbone and thus a lower DR. For example, 10 ppm of a native XG solution is as efficient as 50 ppm of a modified XG solution on %DR (see Fig. 2). At 10 min, the %DR efficiency of modified XG decreased rapidly as result of polymer degradation. The mechanical degradation of the polymeric chains under turbulent flow is a function of diverse factors, such as the molecular weight, temperature, interaction between the polymer and solvent, and molecular structure [29–32]. As shown as Fig. 2, a graph of the efficiency of drag reduction showed that the %DR of pristine XG was higher than modified XG up to 2. This is why the modified XG has more rigidity because of the grafting of octylamine onto XG. The mechanical scission of XG with a rigid functional group became more accelerated compared to pristine XG due to the increased hydrodynamic volume with a rigid functional group, thereby providing a higher shear viscosity [30]. In other words, the conformation of XG, which is accountable for the unusual stability of the polymer, had a lower drag reduction efficacy with its modification.

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