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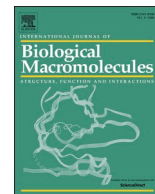
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Eco-friendly sodium alginate for drag reduction in hydraulic fracturing: Experimental and FFT-based dynamic analysis

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ABSTRACT

Hydraulic fracturing is necessary for petroleum engineering, yet synthetic drag-reducing polymers often introduce environmental hazards. In this work we use sodium alginate, a seaweed-originated biopolymer, to address these concerns. Experiments conducted in a smooth microtube show that the eco-friendly additive can reduce the friction to 15 %, and application of the scaling law suggests a drag reduction up to 37 % in a large pipeline. Fast Fourier transform of a newly defined dimensionless pressure drop reveals a spectrum for sodium alginate that is simpler than that of the pure solvent, confirming suppression of turbulent fluctuations through the polymer-turbulence interaction. A ten-minute degradation test indicates that the initial drag reduction of 15 % declines only to 13 %, demonstrating a good anti-shear ability in the drag-reducing flow. Dynamic-light-scattering profiles confirm that chain scission is only partial, underscoring the polymer's structural robustness under the turbulent flow. Last, we use thermodynamic and kinetic, supported by dynamic-light-scattering data, to provide a mechanism of drag reduction by polymers: the structure of sodium alginate resists the high shear rates encountered in turbulent flow, preserving its drag-reducing efficiency. These combined environmental and economic advantages indicate sodium alginate as a promising green additive for hydraulic-fracturing operations, especially under stringent ecological regulations.

1. Introduction

1.1. Environmental challenges of synthetic drag reducers

Currently, even though renewable energies, such as wind, solar and nuclear energy are booming, the petroleum industry still plays the dominant role in the world. In the development of oil and gas, the traditional methods of extraction gradually receive less attention due to their low efficiency, so new methods receive more, and hydraulic fracturing is among these methods. Hydraulic fracturing refers to the introduction of fluids which contain water and several chemicals, to the oil reservoir to extract oil and gas. Unlike conventional methods, hydraulic fracturing is very useful in the development of petroleum and can increase production significantly. However, the use of drag-reducing

polymers in hydraulic fracturing can have significant environmental impacts [1,2].

The environmental implications of hydraulic fracturing are multifaceted. One of the key concerns is water contamination. Hydraulic fracturing can lead to groundwater and surface-water pollution due to spillages and leakages. The process can also alter the deep sub surface's biogeochemical landscape, potentially affecting reservoir sustainability and infrastructure integrity, even the climate, the latter intensifying water contamination issues [3,4]. These environmental challenges underscore the need for more sustainable practices in the application of hydraulic fracturing, particularly in the choice of chemicals used. Besides, a recent study indicates that drag reducers can lose more than the drag-reduction ability within minutes under high shear rates [5].

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1.2. Rationale for sodium alginate

In the application of hydraulic fracturing, several chemicals are needed and drag-reducing polymer is the one among them. During the process of hydraulic fracturing, the flow friction, which is known as the resistance to flow when the fluid flow passes an object due to the interaction between the fluid flow and the surface of the object, exists [6]. This friction can cause energy loss during liquid transportation. To save the energy, researchers attempt to develop different method to reduce the friction, such as the micro-structure of the surfaces, coatings and chemical additives, among which, the most frequently used method is introducing chemical additives, such as polymers, surfactants, bubbles and nano-particles, to the fluid to reduce the resistance, also known as the drag reduction by agents [7]. This method was found by Toms who added polymethyl methacrylate (drag-reducing polymer) in chlorobenzene and found that the pressure drop decreased after the drag-reducing agent was added in a turbulent pipe flow [8].

Since Toms discovered this phenomenon in 1947, numerous works started to focus on this research area due to its huge benefits for hydraulic fracturing [9–13]. However, after researchers published many works, they gradually realized that most synthetic drag-reducing polymers mentioned above would bring environmental problems if they continued to use them in hydraulic fracturing [14–17]. Thus, several other papers used natural bio-based polymers to reduce the friction in the turbulent flow [18].

Bio-based drag-reducing polymers refer to the polymers extracted from either natural resource, such as *aloe vera*, algal and papaya etc., or product of biochemical reactions. Abdulbariet al. extracted a bio-based polymer from *aloe vera* and used a microchannel flow line to test its ability of drag reduction [18–22]. The experimental results indicated that the drag reduction could be up to 60 % when the polymer concentration was at several-hundred-ppm (parts per million) level. To reduce the friction between the ship and sea, Gasljevic and Matthys used a bio-based polymer from a marine microalga and investigated its feasibility as a drag-reducing agent [23]. The maximum drag reduction was 40 % when the concentration was 50 ppm. Rajappan & McKinley investigated the polysaccharide extracted from the mucilage in the research of drag reduction and degradation [24]. It was found that this polysaccharide could reduce the friction in turbulent flow up to 15 %. Campolo et al. found that xanthan gum (XG), a product of fermentation of *xanthomonas campestris*, was a good drag reducer in turbulent flow [25]. The drag reduction varied with the Reynolds number and concentration and could be up to 80 % at 200 ppm. What was more, XG also had a good ability to resist the high shear rate in the turbulent flow so that the drag reduction could be maintained for a long time, which was supported by several papers [26,27]. Besides, DNA (deoxyribonucleic acid) was another type of bio-based drag-reducing polymer, which had been applied in several studies [28–31]. These works suggested that under a low concentration, DNA started to show the excellent drag reduction ability in both pipe flow and rotational flow; besides, DNA also had a good stability in the high shear rate in turbulent flow due to the double-helix structure, so the drag reduction could last longer than the one by flexible linear polymers, such as polyethylene oxide and polyacrylamide.

Besides what is mentioned above, sodium alginate, derived from marine brown algae, is also a biopolymer, which is a good candidate of drag-reducing polymer suggested by Hoyt [32]. However, to our best knowledge, there are few works which investigated the drag reduction by this biopolymer and provided the solid experimental data of drag reduction in turbulent flow. So, in this work, we investigate the property of drag reduction by sodium alginate as follows.

- Objective 1: Quantify sodium alginate's drag-reduction efficiency in microtube turbulent flow and predict its performance in full-scale hydraulic-fracturing pipelines.

- Objective 2: Evaluate the biopolymer's shear-resistance (anti-degradation) and clarify the drag-reduction mechanism via dynamic light-scattering analysis.

2. Experiments

The experiments are performed in a microtube flow apparatus, as shown in Fig. 1. Polymer solution is in a tank; a metering advection pump (2 PB-100001 II, Szweico, China), which flowrate is controlled by a computer (the uncertainty of flowrate by this pump is 1.5 % of the flow rate), is used to transport the polymer solution into a hydraulically smooth microtube (polymethyl-methacrylate microtube, inner diameter 1.85 mm, length of test section 48 cm). To avoid entrance effect, the entrance length is 30 cm (the ratio of entrance length to the inner diameter more than 100 times) [8]. After the entrance length, the pressure drop of polymer solution is measured by a differential pressure transducer (EJA110E, Chongqing Yokogawa, China) in the 48-cm test section. The full range of the sensor is 50 kPa and the uncertainty is 37.5 Pa.

The polymer used in our experiments is sodium alginate purchased from Tianjin Fuchen chemical reagents factory, China. The average molecular weight of the sodium alginate 300,000 g/mol, and its viscosity is 0.02 Pa·s when the concentration is 1 % provided by the manufacture. The monomer length of sodium alginate is 0.515 nm [33]. To prepare the polymer solution, we add the proper amount of biopolymer into deionized water and mix the polymer under a low rotation (60 rotation per minute) speed for 24 h to guarantee that the polymer solution is homogeneous. To measure the size of sodium alginate in the solution, we use dynamic light scattering (Winner 802, Winner particle analysis, China) [34,35]. All experiments including micro-flow test and dynamic light scattering are performed in the room temperature, 25 °C approximately.

3. Results and discussions

To prove the correctness of data provided by this microtube-flow apparatus, we perform a benchmark test using deionized water without polymers as shown in Fig. 2. Fig. 2 shows the measured friction factors and the calculated ones under different Reynolds numbers. The equations for the measured friction factor, calculated friction factors (f , defined in Eq. 1) in hydraulically smooth microtube for laminar (Hagen–Poiseuille equation) and turbulent flow (Blasius equation), and Reynolds number (Re) are in Eqs. 1, 2, 3 and 4 respectively. In these equations, d is the inner diameter of the microtube, m; ΔP is the pressure drop, Pa; l is the length of the test section of microtube, m; ρ is the density of fluid, kg/m³; u is the bulk velocity, m/s; Q is the flowrate, m³/s.

If data provided by the microtube-flow apparatus are correct, the friction factor should follow the Hagen–Poiseuille equation in the laminar flow and Blasius equation in the turbulent flow. From Fig. 2, we can see that when the Reynolds number is less than 1000, the friction

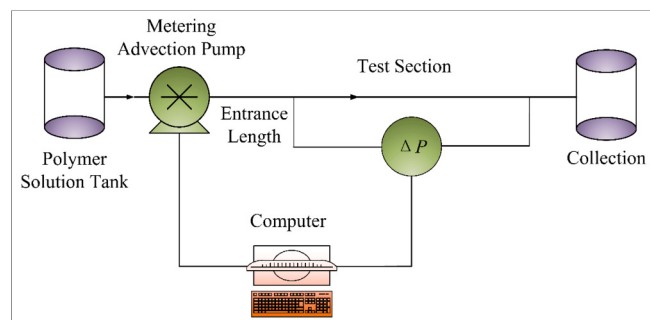


Fig. 1. Microtube-flow apparatus.

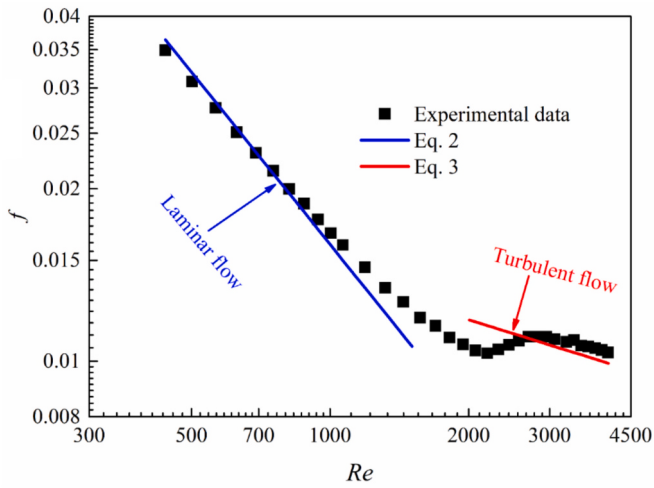


Fig. 2. Benchmark test of microtube flow.

factors follow the Hagen-Poiseuille equation (Eq. 2); when the Reynolds number ranges from 1000 to 2600, the friction factors start to deviate from the Hagen-Poiseuille equation; when the Reynolds number is greater than 2600, the friction factors follow the Blasius equation (Eq. 3). Results above are different from the results in classic theory, where the Reynolds number of transitional flow should be from 2300 to 4000. The difference is due to the high shear rate in microtube turbulent flow. To achieve the same Reynolds number (Eq. 4), we need a higher velocity in a small diameter (microtube) than the one in the large diameter (regular diameter). Under this condition, the shear rate, defined as the $8u/d$ in the small diameter is greater than the one in large diameter. Since the shear rate is higher, the momentum transport in the microtube is more chaotic than the one in the regular diameter. Hence, the friction cannot be predicted by classic equations, and the Reynolds number in the transition regime is different from the one in the classic theory. As shown above, these observations, the relationships between the Reynolds number and friction factor, are fully consistent with the theory of micro-channel transition flows with a small deviation: in laminar flow, the friction factors follow the Hagen-Poiseuille equation; in turbulent flow, the friction factors follow the Blasius equation. Under these conditions, we can confirm that data provided by this microflow apparatus are correct, so the microflow apparatus is reliable and can provide the correct experimental data.

$$f = \frac{d\Delta P}{2l\rho u^2} = \frac{\pi^2 d^5 \Delta P}{32l\rho Q^2} \quad (1)$$

$$f = \frac{16}{Re} \quad (2)$$

$$f = 0.079Re^{-0.25} \quad (3)$$

$$Re = \frac{du\rho}{\mu} \quad (4)$$

Since the micro-flow device can provide the correct data of flow test, we wish to use it to examine whether the biopolymer, sodium alginate can reduce the friction between the inner surface of the microtube and the flow. Fig. 3a shows the pressure drop with and without sodium alginate under different flow rates and relatively low polymer concentration. It is not surprising to see that under low flow rates, the drag-reduction phenomenon does not happen due to the unchanged onset point of drag reduction by polymers. Virk proposed the famous theory of drag reduction by polymers, where Virk mentioned that drag reduction phenomenon could not happen until the onset point was reached, i.e., the transitional Reynolds number between the regime of laminar flow and the turbulent flow [36,37].

This transitional Reynolds number varies in different types of tube and channel flow: for regular-size flow, this Reynolds number is from 2300 to 4000; for micro-size flow, this Reynolds number ranges from 1000 to 2000 as shown [38,39]. Since we want to know the drag reduction property of sodium alginate, we further increase the flow rate (Reynolds number), but the drag-reduction phenomenon is still not obvious as shown in Fig. 3a. Under this circumstance, we examine the pressure drop carefully at high flow rates, and find that when the flow rate is high enough, the pressure drop with sodium alginate is lower than that of water (pure solvent, without sodium alginate) as shown in the partially-enlarged sub-figure in Fig. 3a, which is due to the polymer conformation change in the flow and will be discussed in the following section. Since the drag reduction phenomenon appears, we increase the polymer concentration further to see if the drag-reduction rate can be higher, which is also supported by Virk theoretically [36,37]. In these two works, Virk proposed the maximum drag reduction line (Eq. 6), in which equation, f is the friction factor, defined in Eq. 1. When the concentration is increased, the friction factors decrease, which is manifested in Fig. 3b: Prandtl-Karman line starts to deviate and will be closer to the maximum drag reduction line after the polymer concentration is increased.

$$\frac{1}{\sqrt{f}} = 4\log Re\sqrt{f} - 0.4 \quad (5)$$

$$\frac{1}{\sqrt{f}} = 19\log Re\sqrt{f} - 32.4 \quad (6)$$

The results of further increase in the polymer concentration are

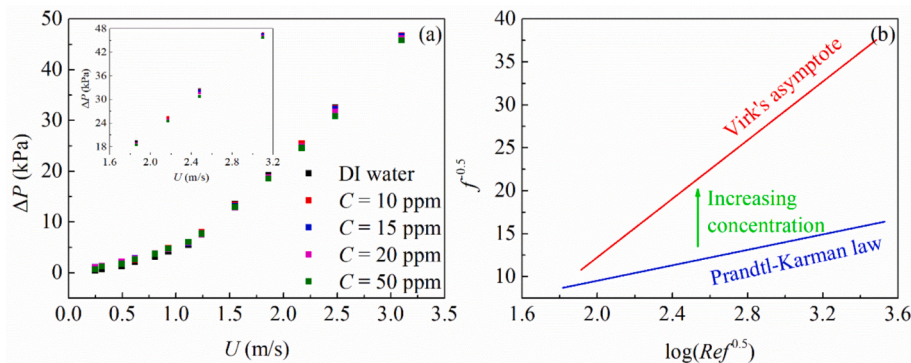


Fig. 3. Effect of sodium-alginate concentration on drag reduction in a hydraulically smooth microtube (a: pressure drop versus flow velocity for polymer concentrations of 0, 10, 15, 20 and 50 weight-based ppm; b: comparison of friction factor in drag-reducing turbulent flow by Prandtl-Karman law and Virk's maximum-drag-reduction line).

shown in Fig. 4. Fig. 4a represents the pressure drop with and without polymers. Obviously, the pressure drop with polymers is lower than the one without, so to some extent, here we can see that the drag-reduction phenomenon happens in the sodium alginate-water system. However, before we confirm the drag reduction happens and analyze drag reduction, we should use the famous criteria to validate our view: in the research of drag reduction by polymers, friction factors should be incorporated by two lines, i.e., Prandtl-Karmen line (Eq. 5) and maximum-drag-reduction line (Eq. 6). In Fig. 3b, we can see that all friction factors are in the area enveloped by these two lines. Under these circumstances, we can confirm that experimental data are correct. In Fig. 4c, we calculate the drag reduction using Eq. 7, in which ΔP_p and ΔP_s represent the pressure drop under the same flow rate with and without polymers. Here we can see that the drag reduction ranges from 1 % to 15 % approximately, and the error bar for the drag reduction is the standard deviations calculated by three parallel experiments. We can see that the drag reduction is similar to the one by epidermal biopolysaccharides [24], lower than the one by guar gum and natural yam mucilage [26,40,41] and higher than the one by DNA [30]. Specifically, at 200-ppm concentration and 0.3 L/min, sodium alginate has a 15 % $\pm$ 1 % drag reduction, about half the 30 %–40 % commonly reported for guar-gum derivatives [26], yet much higher than the 1 %–5 % values documented for DNA under comparable shear conditions [30]. This intermediate performance, combined with its marine origin and low ecotoxicity, indicates sodium alginate as a green drag reducer when maximum drag reduction must be balanced against environmental regulations in hydraulic-fracturing operations. Furthermore, sodium alginate demonstrates a clear economic advantage. Current industrial-grade quotations from [Made-in-China.com](https://www.made-in-china.com) show sodium alginate at 0.50 USD (US dollar) per kilogram, whereas xanthan gum ranges from 1.5 to 2.5 USD per kilogram. Consequently, the unit cost of sodium alginate is much lower. Although its absolute drag-reduction efficiency by sodium alginate is modest, the combination of its lower price and inherent eco-friendly features (marine origin and rapid biodegradability) renders sodium alginate a cost-effective, sustainable choice for

hydraulic-fracturing operations where environmental regulations are strict. From Fig. 4c, we can see that in most cases, the drag reduction increases under a low flow rate and increases until the plateau appears, which is due to the viscosity and elasticity in the drag-reducing flow. When the flow rate is low, the extension of polymer chain is limited, and the viscosity is dominant. Under this condition polymers can merely affect the flow structure, as results, the drag reduction is low. With the increasing flow rate, polymers can be extended further and have a larger influence on the flow structure, so the drag reduction increases. After the plateau, the drag reduction decreases because of the degradation of polymers in the drag-reducing turbulent flow, where high shears exist. Under this high shear rate, the scission of polymer chain happens, which causes the ability of drag reduction to decrease. In the following section, we will provide a more detailed explanation of drag reduction by polymers in turbulent flow. To predict the drag reduction, we use a second-order correlation for the drag reduction, as shown in Eq. 8. Most data can be predicted by this correlation.

$$DR\% = \frac{\Delta P_s - \Delta P_p}{\Delta P_s} \times 100\% \quad (7)$$

$$DR\% = A_0 + A_1Q + A_2C + A_3QC + A_4Q^2 + A_5C^2 \quad (8)$$

Since our aim of this investigation is to find the potential application of sodium alginate in the hydraulic fracturing for petroleum engineering, the drag-reducing performance in large-scale pipe flow is studied. In our current investigation, the scale is small as shown above, so we can use a widely accepted equation to calculate the drag reduction in large-diameter pipe flow, as shown in Eq. 9 [42]. Here we should mention that Eq. 9 is widely used for the scaling issue regarding the drag reduction but is not universal since this equation is still under debate [43]. But we still attempt to use it as a quantitative method since this is widely used and accepted in many works [44–46]. From Eq. 9, we see that if the polymer type and concentration are the same, we can use the lab-scale data to predict the large-scale data, from which, the friction factor in a large scale can be calculated when the Reynolds number, diameter and

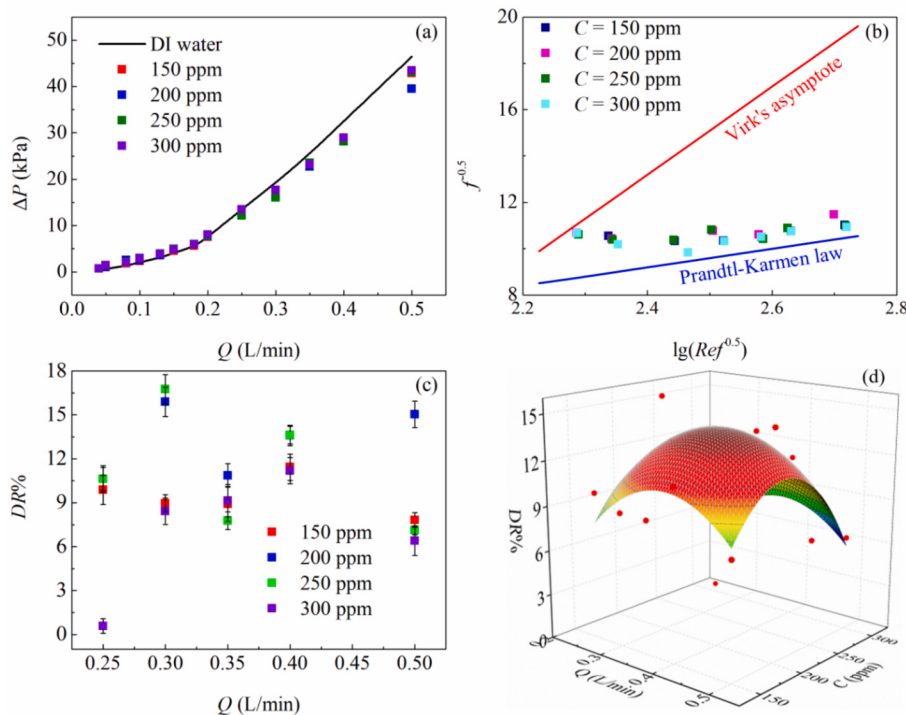


Fig. 4. Drag reduction at elevated sodium-alginate concentrations (150–300 ppm) in microtube turbulent flow (a: pressure drop versus flow rate for polymer solutions and DI water; b: the relationship between friction factors compared by the Prandtl-Karmen law and Virk's maximum-drag-reduction line; c: drag reduction as a function of flow rate and concentration; d: second-order polynomial fit of drag reduction versus flow rate for each concentration).

friction in a small diameter are known. Since the diameter and flowrate in the application of hydraulic fracturing are much larger than the ones in the lab-scale, we set the diameter and flow rate as 0.2 m and 10 m³/min, and the Reynolds number is 8.5×10^5 . These data are based on discussions with Xu Qin, the author in this work, who has been working on a tentative small-scale hydraulic fracturing: in this project, the required flowrate and inner diameter is 0.2 m and 10 m³/min as mentioned above; the predicted drag reduction can be used as an estimation for this tentative project. From our data, the maximum drag reduction appears when the concentration is 250 ppm and flow rate is 0.3 L/min (Reynolds number 3.4×10^3 accordingly), so we combine these data and pre-set data for large scale above to calculate the drag reduction on a large scale. In Fig. 5, we can see that when concentration is 200 ppm, the friction factor for the drag-reducing flow by Eq. 9 is 0.00164, and drag reduction calculated by Eq. 7 is 36.9 %, so the drag-reducing polymer has the potential for hydraulic fracturing. Eq. 9 is used as a rapid method to predict the drag reduction from a small scale to a large scale, and does not consider high salinity, elevated temperature, multicomponent flow, degradation and rheological measurements (e.g., extensional viscosity or relaxation-time spectra) or in-situ viscoelastic characterization, which should be further improved in future works.

$$\frac{Re\sqrt{f}}{d} = Con \quad (9)$$

Previous investigations focus on the static property of drag reduction induced by sodium alginate where no time is involved, so in the following section, we wish to know the dynamic property of drag reduction in which time is involved. First, we focus on how sodium alginate affects the pressure drop in detail. In Fig. 6a, we show the pressure drop with and without sodium alginate versus time. However, we cannot interpret any information from this figure. This is due to the randomness of the turbulent flow. In turbulent flow, the pressure drop versus time is chaotic, so there is no way to explain it. Under this condition, we can transform this pressure drop from time domain to the frequency domain to further interpret the dynamic property of pressure drop when the polymer is added. We can use fast Fourier transform (FFT) to achieve this goal, and here we introduce the transform briefly as shown in Eq. 10, 11 and 12: if we have a series of time-based signals, $x_0, x_1, \dots, x_{n-1}$, we can apply FFT to transform these signals to as follows: X_k are the frequency-domain signals; f_k is the frequency, Hz; Δt is the sampling time, s; Mag_k is the magnitude under a given frequency calculated by the real part (Re_k) and imaginary part (Im_k). So, we can have a series of frequency and magnitude as suggested in our previous work [47].

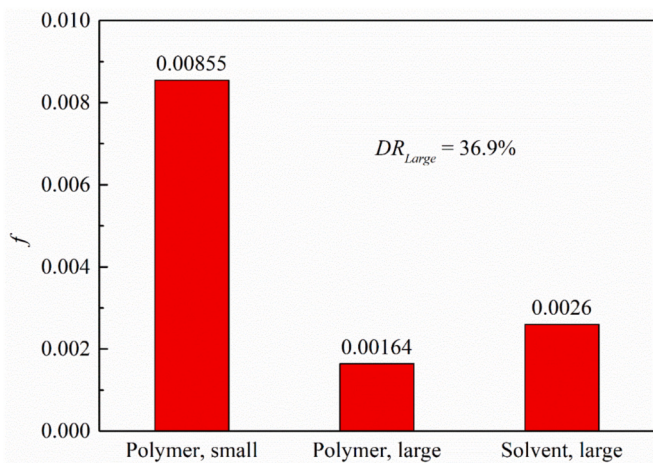


Fig. 5. Calculation of drag reduction and friction factor for larger scale using Eq. 9 (0.3 L/min and 1.85 mm for small diameter; 10 m³/min and 20 cm for large diameter).

$$X_k = \sum_{i=0}^{N-1} x_n e^{-\frac{2\pi k n i}{N}} \quad (10)$$

$$f_k = \frac{k}{N\Delta t} \quad (11)$$

$$Mag_k = \sqrt{Re_k^2 + Im_k^2} \quad (12)$$

In terms of our investigation of drag reduction by sodium alginate, we prepare the time-based signals as follows. ΔP and $\Delta P(t)$ are the average value of pressure drop and time-dependent pressure drop, Pa. Here we define a new function, the dimensionless pressure drop (DPD) as the time-dependent signals in Eq. 13. By using this variable, we can further analyze how sodium alginate affects the pressure drop from the dynamic view.

$$DPD = \frac{\Delta P(t) - \Delta P}{\Delta P} \quad (13)$$

Fig. 6b shows the results of FFT after the time-dependent pressure drop is converted to DPD . From Fig. 6b, we can clearly see the difference: after FFT, the spectrum of DPD of pure solvent (without sodium alginate) has 2 high peaks while the one of sodium alginate is much lower than the one of pure solvent, which shows that some peaks are eliminated by sodium alginate. The reason why sodium alginate can eliminate the peak is the interaction between the flow structure and polymers. As shown in Fig. 6a, there are fluctuations of the pressure drop in the turbulent flow, which indicates that velocity fluctuations also appear in this flow since the pressure drop is related to the velocity. So, we can treat the fluctuation of the pressure drop as the fluctuation of the velocity in the turbulent flow. Besides, in the turbulent flow, velocity fluctuations and vortical structures are two sides of the same coin: vortices are the physical source of fluctuations, while fluctuations are the dynamic manifestation of vortices. Coherent eddies, hairpins and quasi-streamwise vortices, generate streamwise and wall-normal fluctuations via their induced velocity fields; these fluctuations, in turn, intensify or attenuate the vortices through vortex stretching, creating a self-sustaining cycle. Thus, the velocity fluctuation is highly related to the vortex structure in the turbulent flow [48–50].

Since FFT does not change the physical meaning, the spectrum in Fig. 6b for both drag-reducing and non-drag-reducing turbulent flow still represents the fluctuation of the velocity, which also represents the vortical structures. More peaks appearing in Fig. 6b for non-drag-reducing flow means that more velocity fluctuations and vortical structures exist, which indicates the pure turbulent flow without drag reducer is far from the equilibrium state of the velocity, i.e., the average velocity in the microtube flow; when the sodium alginate is added in the turbulent flow, the velocity fluctuations are suppressed as shown in Fig. 6b, which can be regarded as the reduction of intensity of vortex in the drag-reducing turbulent flow. In other words, the sodium alginate extracts the kinetic energy needed to sustain the self-sustaining cycle of vortices. When vortical structures attempt to stretch the polymer chains, polymer chains rebound immediately, preventing further stretching and breakup of the vortices and weakening the vortex-stretching stage. As a result, the eddies are unable to complete their full generation-lifting-breakdown cycle. Consequently, the original peaks in non-drag-reducing turbulent flow vanish and the overall energy shifts downward in the drag-reducing turbulent flow induced by sodium alginate.

We have discussed the drag-reduction ability of sodium alginate, but no degradation, also known as the anti-shear property in turbulent flow, is analyzed. Thus, we still use the microtube-flow apparatus to investigate this characteristic. Degradation is a phenomenon where the polymer loses the ability of drag reduction over a period of time for drag-reducing polymers in the turbulent flow. We perform the degradation test of sodium alginate when the flow rate is 0.5 L/min and 200 ppm. The polymer solution is cycled in the system: it is pumped out from the

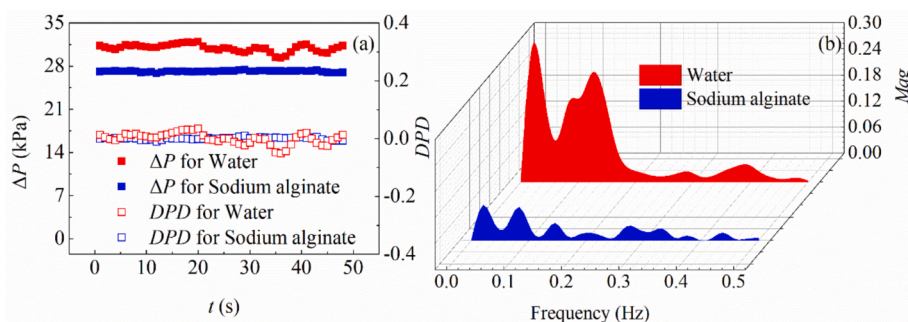


Fig. 6. Dynamic analysis of sodium-alginate drag reduction at 200 ppm and 0.5 L/min (a: time-resolved pressure drops and dimensionless pressure-drop (DPD) signals for pure solvent (black) and alginate solution (red); b: FFT amplitude spectra of DPD over showing the damping of velocity fluctuations by the polymer (sampling frequency 1 Hz, record length 50 s)).

storage tank and returns to the tank again, and the solution will be reused for the degradation test; the total time for degradation is 10 min and the sampling time is 1 min. The polymer solution before and after the degradation is transferred to the test of dynamic light scattering three times without further treatments. All dynamic light scattering tests are performed under the room temperature 25 °C approximately as mentioned before. Besides, the degradation time is also based on the discussion with Xu Qin, the author in this work, who mentioned that the project he has been working on is a tentative small-scale hydraulic fracturing. In this project, the required flowrate and inner diameter are the same as mentioned above (10 m³/h and 20 cm, respectively), and the length of the pipeline is 50 m. The residence time for the fracturing fluid under this circumstance is 9.4 min, close to 10 min approximately. Thus, we choose 10 min as the degradation time. The results are shown in Fig. 7a. From this figure, we can see that drag reduction at the initial state is 15 % approximately; after 1 min, the drag reduction decreases to 13 % and can maintain for 9 min approximately. Thus, sodium alginate exhibits minimal degradation since there is no huge decline of drag reduction over this 10-min test. Hence, it indicates that the sodium alginate has a good ability to resist the high shear rate in the turbulent flow. The reason why sodium alginate has a good ability of anti-shear in the drag-reducing flow will be explained in the following section. This good anti-degradation property is good for hydraulic fracturing since the pipeline for hydraulic fracturing is very long. During the long-distance

travel, the residence time is also very long for polymers, so the polymers tend to be degraded over this period. If the polymer chain is broken, the drag reduction will be lower, and the result of hydraulic fracturing will not be as good as expected. The drag reduction of sodium alginate can be maintained for a relatively long period of time, so the performance of sodium alginate can be better than the one by other polymers which are easily degraded, such as PEO (polyethylene oxide) and PAM (polyacrylamide).

In the previous sections, we have two unexploited questions: how the conformation of polymer affects the drag reduction, and the degradation of polymer in turbulent flow. In fact, we can use our previous theory to interpret these two questions. Our previous works have already established a theory for polymers using thermodynamics and kinetics as shown in Fig. 7b [51,52]. At the initial stage of sodium alginate, the polymer is in the coiled state under a shear-free condition; if the shear rate is strong enough to stretch the polymer chain, the polymer chain is elongated and can interact with the structure of turbulent flow, i.e., the vortex; this process is reversible: the stretched polymer will be in the coiled state again after the polymer chain is elongated, which process can be regarded as the equilibrium state of thermodynamics. Also, the drag reduction also happens in the stage: if the relaxation time (the time describing the transition between the coiled state and stretched state) is long, the polymer can have enough time to interact with the vortex, and the drag reduction can be high. However, this reversible state is not

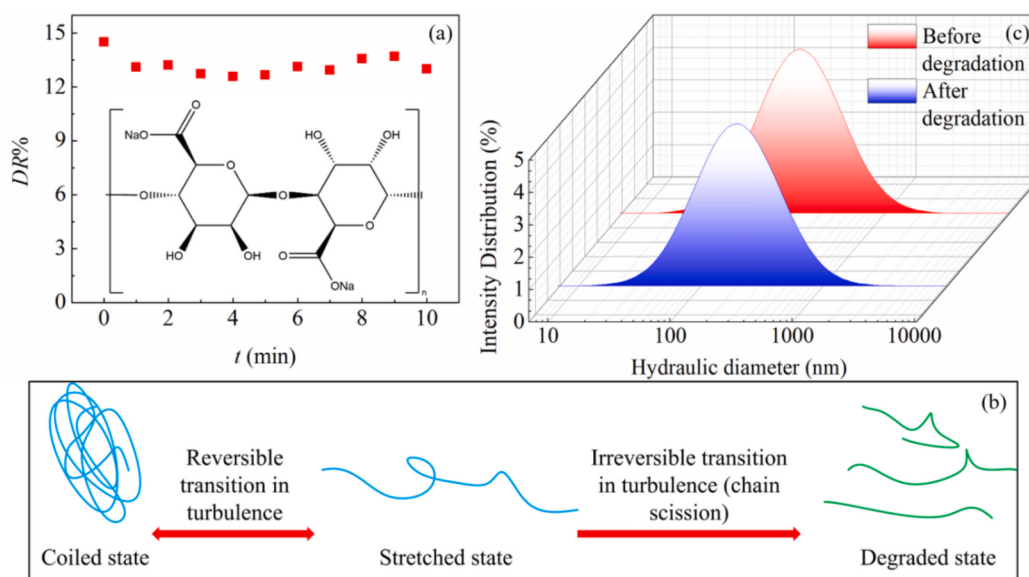


Fig. 7. Degradation test and hydraulic diameter measured by dynamic light scattering and mechanism of drag reduction by polymers (a: degradation test of sodium alginate when $Q = 0.5$ L/min and $C = 200$ ppm in 10 min; b: schematic of drag reduction by polymers in turbulent flow; c: hydraulic diameter of sodium alginate by DLS when $C = 200$ ppm before and after 10-min degradation test).

permanent due to the high shear rate which stretches the polymer chain: since the velocity distribution in the channel or rotational flow is not evenly, i.e., in the near-wall regime, the velocity is small, but the gradient is large, also known as the high shear rate; when the polymer is in this regime, it can be expected that the polymer chain is cut by this high shear rate, and the degradation of polymer happens; so the drag reduction decreases which should be treated as the dynamic process since the polymer-chain scission is irreversible. While the coil-stretch transition is used to explain the drag reduction, the present study does not include direct rheological measurements such as extensional viscosity or relaxation-time spectra. Consequently, the proposed mechanism remains qualitative and awaits future validation through the rheometry or in-situ viscoelastic characterization.

The sodium alginate used in this work has a rigid structure as shown in Fig. 7a, which contains two hexatomic-ring structures, so this polymer can resist the high shear rate in the turbulent flow to some extent unlike the flexible polymers such as PEO and PAM mentioned before, and this integral structure is helpful to maintain the drag reduction in turbulent flow: as stated above, the 13 % drag-reduction rate can be maintained for 10 min. Even though this polymer is rigid, the high shear rate still can cause degradation as shown in Fig. 7b, where we use dynamic light scattering (DLS) to investigate the structure of sodium alginate before and after the degradation test. This method can provide the distribution and the average size (hydraulic diameter) of polymers in the solution, so we can determine these two parameters mentioned above and use them to examine whether the sodium alginate degrades or not in the turbulent flow.

The size distribution of polymers by dynamic light scattering can affect the drag reduction [53,54]: larger size detected by dynamic light scattering accelerates the onset of drag reduction. During the measurement of dynamic light scattering, the decay rate of the dynamic light scattering autocorrelation function tracks the interaction between polymers and vortical structures: slower decay (larger size) corresponds to greater suppression of turbulence, while faster decay (smaller size) reduces the drag-reducing effect. Thus, the breadth and peak position measured by dynamic light scattering are direct indicators of the attainable drag reduction.

Two profiles are shown in Fig. 7c, one for the initial sodium alginate (before the degradation test, red) and the other for the one after the degradation test (blue). We perform each test three times, i.e., before or after degradation. Since in each experiment, the profiles are very similar and overlapped, so we show one profile for each test. From the results, the average size of the sodium alginate before and after the degradation test are 510 and 507 nm respectively (the detailed results in three parallel experiments as follows: 510, 510 and 509 nm for before degradation; 508, 507 and 507 nm for after degradation). Here we use one-way ANOVA (Analysis of Variance) to further investigate whether the average size before and after the degradation test remains the same statistically: by one-way ANOVA, the *P* value is 0.0078, less than 0.05, which indicates that the average size before and after degradation are different statistically. Thus, we can confirm that degradation truly happened. While the difference is not obvious, from 510 nm to 507 nm, the relative deviation is less than 1 %, which indicates that only partial degradation happens. Also, the two profiles are similar, so we can further confirm that only partial degradation happens. Partial polymer degradation may be related to the non-uniform distribution of macromolecules in the microtube flow [55,56]: the high shear rate is in the near-wall regime, so it is possible that polymers in this area are inclined to be degraded by the high shear rate; in the regime far from the near-wall regime, the shear rate is not that high, so the polymers may not be degraded by the lower shear rate. Under this circumstance, only partial degradation happens. A previous study showed that, for flexible polymers, degradations happened completely, and the relative deviation of size measured by DLS could be reduced by 10 % to 50 % depending on the degradation time (2–6 min) [57].

In the current study, we perform a comprehensive analysis of the

application of sodium alginate, a naturally derived biopolymer for the hydraulic fracturing within the petroleum industry. Our findings indicate that sodium alginate exhibits a good characteristic as an eco-friendly additive, its efficacy in terms of drag reduction compared with synthetic drag-reducing polymers remains low. These results align with industry demands for eco-friendly chemicals, positioning sodium alginate as a pragmatic green alternative [58]. The limitation in its performance underscores the necessity for further research and development. Future investigations should be directed towards the preparation of novel bio-based drag-reducing agents that not only minimize environmental impact but also increase drag-reduction efficiency. The ideal biopolymer should be capable of achieving a higher drag-reduction rate while maintaining or even enhancing its eco-friendly features [59]. Besides, all tests are conducted at 25 °C, whereas field temperatures commonly reach 40–60 °C. Future work also should therefore evaluate the thermal stability of sodium alginate under representative down-hole conditions.

4. Conclusion

In this work, we use sodium alginate as the drag-reducing agent to reduce the friction in microtube flow. Experimental results show that sodium alginate can reduce the flow resistance under several conditions and can resist the high shear rate in turbulent flow, both of which indicate that this biopolymer has the potential for hydraulic fracturing in the development of oil and gas. Besides, we also use FFT to analyze the spectrum of newly defined parameter, *DPD*. FFT results represent that the spectrum of sodium alginate is much simpler than the one of pure solvent due to the interaction of sodium alginate and turbulent flow. Besides, we also propose the mechanism of drag reduction in turbulent flow by biopolymers, validating by the degradation test and results by dynamic light scattering. The combined environmental and economic advantages position the sodium alginate as a promising green additive for hydraulic-fracturing operations, especially in regions with stringent ecological regulations.

Nomenclatures

Symbol/Abbreviation	Definition	Unit
<i>C</i>	Polymer concentration	ppm
<i>d</i>	Inner diameter of microtube	m
<i>DPD</i>	Dimensionless pressure drop	–
<i>f</i>	Friction factor	–
FFT	Fast Fourier transform	–
<i>l</i>	Length of test section	m
<i>Q</i>	Volumetric flow rate	m ³ ·s ^{−1}
<i>Re</i>	Reynolds number	–
SA	Sodium alginate	–
<i>u</i>	Bulk velocity	m·s ^{−1}
XG	Xanthan gum	–
ΔP	Pressure drop	Pa
ρ	Fluid density	kg·m ^{−3}
$\dot{\gamma}$	Shear rate	s ^{−1}
μ	Dynamic viscosity	Pa·s

CRedit authorship contribution statement

Xin Zhang: Writing – original draft, Methodology, Conceptualization. **Zhensong Cheng:** Software, Investigation. **Xiaodong Dai:** Resources, Funding acquisition. **Yanping Xin:** Data curation. **Kun Liu:** Formal analysis. **Mingzhen Lin:** Methodology. **Yue Liang:** Validation. **Yihu Xiao:** Investigation. **Yuan Lu:** Validation. **Guoxing Zhang:** Investigation. **Xu Qin:** Formal analysis. **Jianhua Zhang:** Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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