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Experimental Study on the Rheological Property of Aqueous Solution of Polyacrylamide†

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With polyacrylamide solution as experimental subject, the paper reports the effect of mass concentration, shear rate and temperature on the rheological properties of polyacrylamide solution. Experimental results indicated that the apparent viscosity of polyacrylamide solution increased linearly with linear increasing concentration at concentration of 0.5-2.0 g/L; the apparent viscosity decreased with the increasing of shear rate and temperature. With the changing of temperature and concentration, the flow behaviour index (n) and consistency coefficient (K) behave similar to power-law fluid. Considering the combined effects of temperature and concentration, the experiment fitted an equation of the apparent viscosity with regard of concentration and temperature.

Keywords: Polyacrylamide aqueous solution, Rheological property, Apparent viscosity.

INTRODUCTION

Polyacrylamide (PAM) aqueous solution is a typical non-Newtonian fluid. Most of applications of PAM have been made in the format of aqueous solution. Polyacrylamide is soluble at any concentration, temperature and pH value in water. Polyacrylamide is water-soluble high molecular polymer, which is not soluble in most of organic solvents. Solid phase PAM has feature of moisture absorption, flocculability, gumminess, friction reducible and thickening property. It has been widely applied in petroleum industry, oil production, drilling fluids, waste mud treatment, prevention of water channeling, reduction of friction reduction, enhancement of oil recovery, tertiary oil recover and water treatment. In this paper PAM was mainly used as flocculant for suspended particles, coarser, high concentration, particles with positive charge, neutral water or alkaline wastewater. Because the anionic polyacrylamide molecule chain contained a certain amount of polar groups which can absorb suspended solid particles in water, which formed large floccules between particle bridges. So it accelerated the sedimentation of particles in suspension, speeding up solution clarification obviously and promoting the effect of filtering. It has been widely used in the chemical industry wastewater and waste fluid treatment, municipal sewage treatment. The applications of PAM in petroleum include refinery water treatment and recycling of treatment oil and in oilfield wastewater treatment.. Some experiments on studying rheological property of PAM solution have been carried out in previous works[1-4], but different type of PAM solution has its own rheological properties. For non-Newtonian fluid, it is difficult to reflect its property using the analytical form of rheological equation. τ-γ flow curve is usually used to describe the rheological properties[5,6]. For power-law fluid, one of simplest non-Newtonian fluids, the relationship between shear stress τ and shear rate γ can be described by equation. This paper used a rotary rheometer to measure apparent viscosity and shear stress of PAM solution at different concentration. Through experiment, this study obtained the effect of concentration, temperature and shear rate on apparent viscosity, consistency coefficient and flow behaviour index. The results were used to further obtain the value of consistency coefficient (K) and flow behaviour index (n) and corresponding PAM rheological constitutive equation, which can be used to study non-isothermal flow problems of PAM solution and other similar power-law fluids.

EXPERIMENTAL

Experimental samples and instruments: During the polyacrylamide manufacturing process, high drying temperature and long drying time made the product partially branched or lightly crosslinked, which extended the dissolution time. The experiment samples of polyacrylamide were obtained from HYMO joint-stock company, Japan. It molecular weight is 10 million and degree of hydrolysis is 20-30 %. The largest

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shareholder of HYMO is Mitsubishi Chemical Corporation and the product annual output is 50,000 tons, ranking top two in Japan. The weight was measured with a high accuracy analytical balance (METTLER TOLEDO). A German IKA magnetic stirrer was used to accurate control the stirring temperature. The used rheometer was Austrian Anton PaarPhysica MCR301 advanced rheometer, which is modeled and smart. It is equipped with an accurate temperature control system, which has temperature controlling range -30 ℃ to +190 ℃. Meanwhile, its rotational speed can reach 10^{-7}-3000 rpm. The self- rheological controlling system is featured excellent compatibility, reliability and flexibility. It has multiple application modes including flow and viscosity curve, temperature test and transit test, which can fully fulfill the requirement of the experiment.

Experimental method: Within the 0.5-2.0 g/L mass concentration, PAM solution is dilute solution. The different weighs of polyacrylamide particles were measured with analytical balance. Tap water with pH of 6 was used as solvent. The concentration of prepared solution was 0.5, 1.0, 1.5 and 2.0 g/L, respectively. Because large amounts of hydrogen bond and molecular chain entanglement exist in polyacrylamide, it is difficult to dissolve. Stirring can accelerate the molecular diffusion rate and dissociate the hydrogen bond, making molecular chain tangles untied. This experiment used a magnetic stirrer to help the dissolving process. In order to avoid formation of flocculation during preparing solution and cause uneven sample, the speed of application was required to be even and the stirring speed was controlled to be lower than 200 rpm, which can prevent unnecessary bubbles in solution and degradation of the polymer. The stirring time was set to be 1-2 h according to the concentration. The solution was standing for 12-24 h allowing adequate time for dissolving of the polymer to obtain the even samples. The viscosity of prepared sample solution was measured with rheometer for different concentrations, shear rates and temperatures. The temperature interval was set to be 10 ℃ in the range of 20-50 ℃[7]; flow behaviour index (n) and consistency coefficient (K) was determined according to the measured data.

RESULTS AND DISCUSSION

Effect of mass concentration on apparent viscosity of solution of polyacrylamide: Mass concentration is one of important factors that affect apparent viscosity of polymer solution. Therefore, the measurements were carried out for polyacrylamide solution at mass concentration of 0.5, 1.0, 1.5 and 2.0 g/L.

Fig. 1 shows Rheo-curve of apparent viscosity and shear rate of polyacrylamide solution at different concentrations at 30 ℃. It can be seen from Fig. 1 that at the same condition of temperature and shear rate, apparent viscosity of polyacrylamide solution was high at high concentration during entire experiment.

From Fig. 1, significantly difference of apparent viscosity between low and high concentration polymer solution can also be observed, especially at low shear rate region. With increase of mass concentration, the amplitude of increment of corresponding apparent viscosity also increased. This phenomenon was particularly evident for high concentration polymer solution.

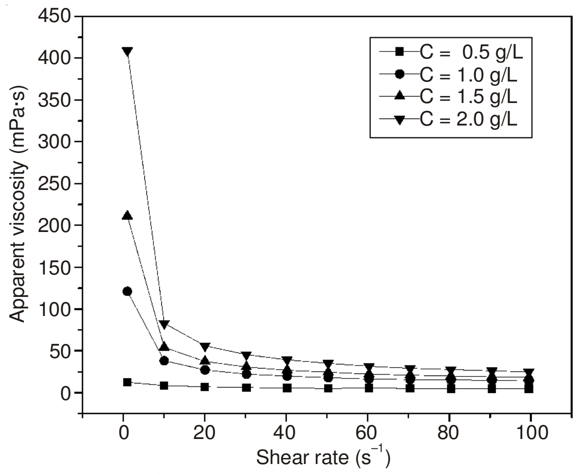

Fig. 1. Rheo-curve for the polyacrylamide solution at different concentrations at 30 ℃

The apparent viscosity of 0.5 g/L solution of PAM is lower than that of other high concentration PAM solutions. It belongs to low concentration solution and the number of hydrogen bond and molecular chain entanglement is less than that of other high concentration PAM solutions. Network structure was easy to be destroyed. With the increase of shear rate, the apparent viscosity decreased. It is not difficult to find out that the variation of apparent viscosity of PAM solution was not obvious in the entire shear rate range for the lowest mass concentration of 0.5 g/L. The feature of non-Newtonian is not strong and it behaved similarly to Newtonian fluid. However, with increase of the mass concentration, the Rheo-curve became more obvious and pseudo-plastic feature increased.

Effect of shear rate on apparent viscosity: According to experimental plan, the shear rate was increased continuously from 1.01 to 100 s^{-1} and apparent viscosity of polyacrylamide solution was measured at different mass concentrations. According to non-Newtonian fluid, taking logarithm would obtain, which demonstrates linear correlation between viscosity and shear rate.

Fig. 2 shows the logarithm relation between shear rate and apparent viscosity of PAM solution at different concentrations and temperature of 40 ℃. All curves showed linear relation except for the lowest concentration curve and illustrated the power law flow characteristics.

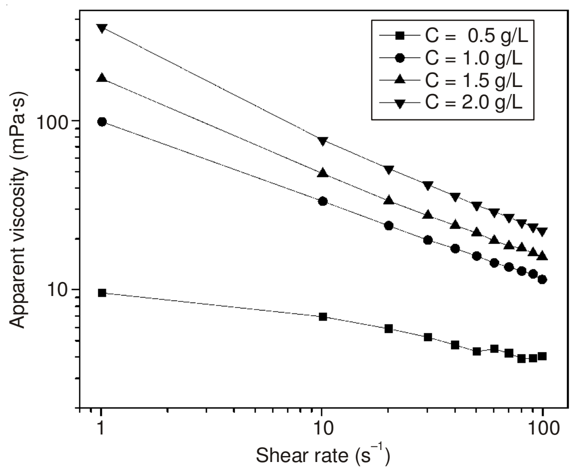

Fig. 2. Plot of logarithm of the shear rate *versus* the apparent viscosity at 40 ℃

From Fig. 2 it illustrated that apparent viscosity decreased with increase of shear rate for polyacrylamide solution at all mass concentrations. The amplitude of this decrease was more significant for high concentration solution. Observing polymer solution with concentration of 0.5 and 2.0 g/L found that the effect of shear rate was relatively weak for low concentration of PAM solution compared to that for high concentration solution. The decrease occurred in the initial low shear rate range, with the continuous increase of shear rate, apparent viscosity appeared flattened. For the solution with concentration of 2 g/L, the apparent viscosity decreased significantly at low shear rate and the decreasing amplitude gradually decreased with increase of shear rate.

Fig. 3 illustrates the relation between shear rate and shear stress of polyacrylamide solution at 30 ºC and different mass concentrations. Fig. 3 showed that the shear stress of polymer solution increased with the increase of shear rate in experimental range of concentration. The flow curve also agreed with pseudo-plastic flow of non-Newtonian fluid. From constitutive equation one can derive, non-linear regression can be performed to the measured τ and γ. This experiment applied least square fitting to shear rate and apparent viscosity and obtained corresponding K and n (Tables 1 and 2).

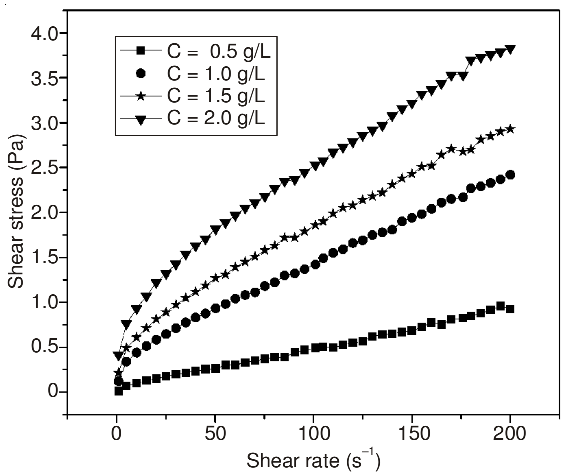

Fig. 3. Plot of stress *versus* shear rate with different mass concentrations at 30 ºC

TABLE-1
VALUES OF CONSISTENCY INDEX OF SOLUTIONS OF POLYACRYLAMIDE WITH DIFFERENT MASS CONCENTRATIONS AND TEMPERATURES

T (K)	K (mPa s^n)			
	C = 0.5 g/L	C = 1.0 g/L	C = 1.5 g/L	C = 2.0 g/L
293	16.413	132.342	240.102	458.453
303	12.987	120.779	210.073	407.424
313	10.165	99.151	176.833	357.106
323	8.282	80.050	147.401	315.717

From Tables 1 and 2, at the same temperature, K continued to increase with the variation of solution concentration from low to high, whereas n decreased with the increase of concentration. This results is consistent with the result of Yang[8]. Compared with the results of Ye Xi[9], it was found that the rheological trend was similar and the apparent viscosity of this experiment PAM solution was larger. Meanwhile, at certain

TABLE-2
VALUES OF FLOW BEHAVIOUR INDEX OF SOLUTIONS OF POLYACRYLAMIDE WITH DIFFERENT MASS CONCENTRATIONS AND TEMPERATURES

T (K)	n			
	C = 0.5 g/L	C = 1.0 g/L	C = 1.5 g/L	C = 2.0 g/L
293	0.7525	0.5142	0.4301	0.3374
303	0.7767	0.5108	0.4376	0.3479
313	0.7978	0.5301	0.4562	0.3641
323	0.8065	0.5635	0.4817	0.3818

mass concentration, n increased with increase of the temperature, whereas K decreased with the increase of temperature. K is consistency index, which reflects the viscosity of the solution. The solution is more viscous with larger K value and it has been validated by experimental result. The value of n for all solution was smaller than 1, which belonged to pseudo-plastic fluid. The larger the concentration, the smaller the value of n. The deviation was increased from the value of n for Newtonian fluid, *i.e.*, n = 1, which indicated that non-Newtonian behaviour of the polymer solution was more obvious.

Effect of temperature on apparent viscosity: The experiment measured the mass concentration of the solution was 0.5, 1.0, 1.5 and 2.0 g/L. We also measured apparent viscosity for these polyacrylamide solution at 20, 30, 40 and 50 ºC. Fig. 4 shows flow curve of apparent viscosity and shear rate for polymer solution at mass concentration of 1.5 g/L. Form Fig. 4 it can be seen that, for same concentration PAM solution, apparent viscosity decreased with increase of temperature. The dependency of viscosity to temperature is usually described using Arrhenius equation[10],

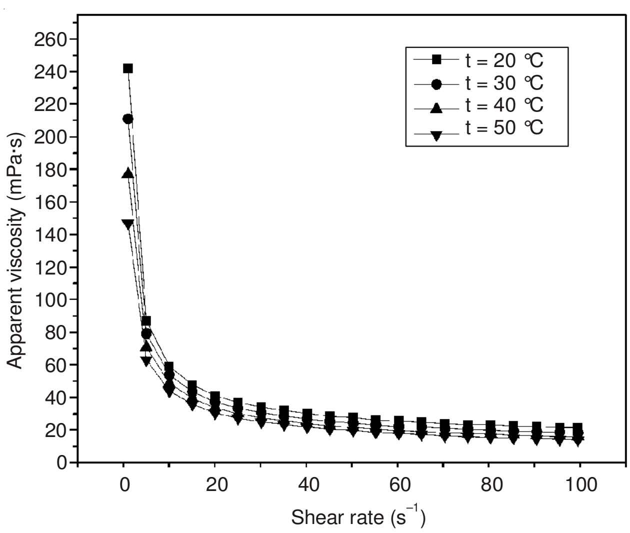

Fig. 4. Function of shear rate and apparent viscosity for 1.5 g/L PAM solution at different temperatures

$$\eta = Ae^{\frac{\Delta E_a}{RT}} \quad (1)$$

where ΔE_a is flow activation energy, A is constant. Large ΔE_a indicates strong temperature dependency.

Relation equation of apparent viscosity with temperature and concentration: To simplify the experimental research application, comprehensively considering the combined effect of temperature and concentration on apparent viscosity and fitting a relationship of apparent viscosity of polyacrylamide solution with respect to both temperature and concentration are very essential.

According to above result, combining the research finding of Yang[11], the correlation of exponential model is higher than that of power law model[11-14]. Therefore, the study used following equations:

$$\eta = K\gamma^{n-1}$$

$$n = A_1 \exp\left(a_1 C - \frac{E_a}{RT}\right)$$

$$K = B_1 \exp\left(b_1 C + \frac{E_a'}{RT}\right) \quad (2)$$

We applied eqn. 2 to obtain logarithm of measured data and performed multiple linear regression. The fitting result showed good correlation of this equation, with correlation coefficient was 0.946 and 0.982. Therefore, the equation for description of the rheological property of polyacrylamide aqueous solution in experimental temperature and concentration range, can be written as

$$\eta = K\gamma^{n-1}$$

$$n = 2.664955 \exp\left(-0.50266C - \frac{2629.951}{RT}\right)$$

$$K = 0.030807 \exp\left(2.21324C + \frac{13527.96}{RT}\right) \quad (3)$$

where C is mass concentration in unit of g/L, R is gas constant and T is absolute temperature.

To examine the reliability of eqn. 3, measured data was compared to the calculated result using eqn. 3 and the comparison was shown in Fig. 5. Fig. 5 showed linear correlation between measured data and theoretical calculation result, demonstrating good agreement. Therefore it can be concluded that eqn. 3 was reliable for relation of apparent viscosity and temperature and concentration. In the range of experimental conditions, it can accurately describe the rheological property of aqueous solution of polyacrylamide.

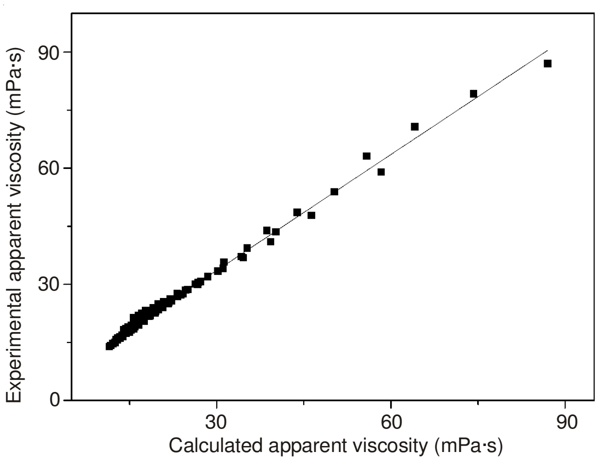

Fig. 5. Relationship between calculated apparent viscosity and experimental apparent viscosity

Conclusion

• The polyacrylamide solution in this study treated as pseudo-plastic fluid as. Apparent viscosity decreased with increase of shear rate, showing diluted shear phenomenon. Furthermore, when shear rate was large enough, the apparent viscosity would not decrease.

• Under certain temperature and shear rate, apparent viscosity of polyacrylamide solution increased with increase of concentration. The amplitude of increment also increased with increase of concentration and this increment demonstrated a linear trend. Flow behaviour index (n) decreased with increase of concentration. Consistency index (K) increased with increase of concentration and non-Newtonian feature increased.

• At the same concentration, apparent viscosity of polymer solution decreased gradually with increase of temperature. The value of n became larger and larger, whereas K became smaller and smaller, demonstrating the dependency to temperature.

• This paper also investigated the combination effect of concentration and temperature on apparent viscosity. Within the measured temperature range of 20-50 ºC and the mass concentration range of 0.5-2.0 g/L, this study fitted the relationship between apparent viscosity and both concentration and temperature. Well description of rheological property of polyacrylamide solution was shown using this equation.

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