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Rheological Behavior of Polymeric Drilling Fluids: A Comparative Study of Carboxymethyl Cellulose, Polyanionic Cellulose, Hydroxyethyl Cellulose, and Xanthan Gum.

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Abstract

The use of water-soluble polymers has increased significantly in many industrial fields. Cellulosic polymers such as carboxymethyl cellulose high viscosity (CMC HV), polyanionic cellulose (PAC R), and hydroxyethyl cellulose (HEC), along with the biopolymer xanthan gum (XG), are widely used to increase the viscosity of drilling fluids. This study analyzes and compares the rheological behavior of aqueous solutions based on these viscosifiers at a concentration of 0.4 wt. %. The results showed that only the XG solution exhibited viscoplastic behavior. While the CMC HV, PAC, and HEC solutions exhibited pseudoplastic (shear-thinning) behavior, the HEC solution displayed a high degree of shear thinning. The apparent viscosities measured at a shear rate of 10 1/s were 0.526, 0.268, 0.153, and 0.050 Pa.s for XG, HEC, PAC and CMC HV, respectively. At a high shear rate of 500 1/s, the apparent viscosities were comparable: 0.026 Pa.s for XG, 0.054 Pa.s for HEC, 0.029 Pa.s for PAC, and 0.025 Pa.s for CMC.

Keywords: *Biopolymer drilling fluids, Rheological models, Apparent viscosity, yield stress.*

I. Introduction

Water-soluble polymers (WSPs), whether natural or synthetic, dissolve in water and modify the physical properties of aqueous systems. WSPs are often used in small quantities as thickeners, stabilizers, and flocculants in numerous applications [1]. For example, polyacrylamides are commonly used as flocculants in wastewater treatment and in the oil recovery industry, while polyacrylic acids are used as cement modifiers [2]. Polymers such as polyethylene glycol, polypropylene glycol and polyvinylpyrrolidone are used in the pharmaceutical industry [3] and as emulsion stabilizers and thickeners [4, 5].

Polymer viscosifiers are widely used in water-based drilling fluid (WBDF) and for enhanced oil recovery (EOR) operation to increase the viscosity of fluids. Various types, such as methylcellulose, carboxymethyl cellulose (CMC), Polyanionic cellulose (PAC) hydroxyethyl cellulose (HEC), partially hydrolyzed polyacrylamide (PHPA) hydroxypropyl methyl cellulose, and xanthan gum (XG) [6] are used in the oil industry. In WBDFs, WSPs are often used to replace bentonite or as a supplement to improve fluid viscosity. For instance, XG and HEC have been used to improve the rheological properties of WBDFs [7]. Ouaer and Gareche [8], Benyounes et al. [9] investigated the effect of HEC concentration on the rheological properties of WBDF. In another study, de Aguiar et al. [10] confirmed the potential use of polymer (PHPA) as viscosifying agent for EOR.

Polymeric viscosifiers differ from each other in their origin (natural, modified, synthetic), ionic nature (charged and uncharged), molecular mass, structure (linear, branched, cross-linked), etc. The molecular mass influences properties like viscosity, chain entanglement, and solution stability.

Structurally, polymers are classified as linear, branched, or cross-linked networks [11]. Linear polymers, which consist of a single chain of monomers, include natural rubber, elastomers, CMC, PHPA, and HEC, and are often characterized by high elasticity [12-14]. A chain of branched-chain links has side branches, for example, amylopectin, starch, and xanthan gum [15].

Even though the viscosifying polymers have high molecular weight, many differences exist: some are linear, others are branched, some are charged, others are not charged, some have functional groups, others do not have. Previous investigations into the rheology of WSP solutions have not included a direct comparison of the key polymers used in

WBDFs. Therefore, to facilitate appropriate polymer selection and to better understand the performance of WSPs in drilling fluid applications, a rheological characterization is necessary. This investigation, therefore, aims to study the effect of water-soluble polymer type (XG, HEC, PAC R and CMC HV) on the rheological properties. Then, a comparative analysis between these different polymers will be established.

II. Material and methods

Water-soluble polymers

Xanthan gum (XG) is a biopolymer produced by the bacterium Xanthomonas campestris through a fermentation process. XG is a water-soluble polymer with high molecular weight, and serves as a thickener, stabilizer, and viscosity modifier.

Hydroxyethyl cellulose (HEC) is a synthetic polymer derived from cellulose, created by chemically reacting insoluble cellulose with ethylene oxide. HEC is as a water-soluble polymer derived from cellulose, used as a binder and thickening agent. HEC is a biodegradable polymer, known for its biocompatibility and non-toxicity. The HEC used in this study is a commercial product, which is used by the oil companies.

Both carboxymethyl cellulose high viscosity (CMC HV) and Polyanionic Cellulose premium-grade (PAC R) are modified polymers derived from cellulose. CMC HV and PAC R are obtained by treating the insoluble cellulose with chloroacetic acid, to introduce carboxymethyl groups onto the cellulose backbone. Both of cellulose-derived products differ from each other in their degree of modification and specific applications. PAC R is an anionic cellulose ether with a high degree of polymerization and substitution. The Table 1 provides information on the properties of polymers, such as their physical and chemical characteristics.

Preparation and measurement

WSPs solutions were prepared according to the method described in a previous study [16]. Solutions were prepared by gradually adding biopolymer powder to water while agitating to prevent the formation clumps and ensure proper dispersion. According to the WSP type, four suspensions were prepared with a concentration of 0.4 wt. %; this concentration is commonly used to formulate water-based fluid formulations [17].

The rheological measurements were performed at 20°C using a controlled stress rheometer AR 2000 (Figure 2) from TA instruments using the concentric cylinder measuring system. The rheometer equipped with Couette geometry measures fluid properties by rotating the bob coaxial cylinder, while the cup cylinder remains stationary.

In order to avoid any memory effect, the sample was pre-sheared at 100 1/s for 60 s after loading in the measuring device. The sample was then kept at rest for 600 s before measurements to allow the material to recover its initial structure. The imposed shear rate ranges from 0 to 700 1/s. A ramp of 0.2 Pa to 300 Pa during 600 s has been applied.

Rheological models

The Herschel-Bulkley (H-B) model is a generalized equation characterized by three parameters: consistency (k), flow index (n), and yield shear stress (τ_0). H-B fluid requires an initial yield stress to be exceeded before flow starts. Classically, the model of Herschel-Bulkley was used to fit the shear rate dependence of the shear stress of WBDF [7, 18]. Moreover, this model can well simulate the rheological properties of cement paste [19]. Therefore, experimental data were fitted to the classical model of Herschel–Bulkley (Eq. 1):

$$\tau = \tau_0 + \mathrm{K}.\dot{\gamma}^n \qquad \text{(Eq. 1)}$$

where τ_0 is the yield stress, if τ_0 is equal to 0, this equation is equivalent to the Power Law model. The consistency index is designed by K letter, the higher value represents the more viscous material, and n is the flow behavior index (power law exponent) giving a measure of the pseudoplasticity. this parameter (n) indicates a degree of dependence of viscosity on the shear rate.

Table 1. Physicochemical properties of the water-soluble polymers studied.

Characteristics	Xanthan gum	HEC	PAC R	CMC HV
Origin	natural	Modified	Modified	Modified
Structure	branched	linear	linear	linear
Nature	charged	uncharged	charged	charged
R groups	$-COO^-$ $-COCH_3$	H/CH_2CH_2OH	H/CH_2COONa	H/CH_2COONa
Stiffness of chains	semi-rigid	rigid	semi-rigid	flexible
molecular weight [g/mol]	1×10^6 to 2×10^6	$9.5x10^5$	$7x10^5$	$7x10^5$
functional groups	acetyl groups carboxylic groups	hydroxyethyl	carboxymethyl groups	carboxymethyl groups
Solubility	Water	Water	Water	Water
degree of substitution	/	1	07 – 1.2	0.65 - 0.75

pH (1 % solution)	6-7	6-8.5	6.0 to 8.0	6.0 - 8.0

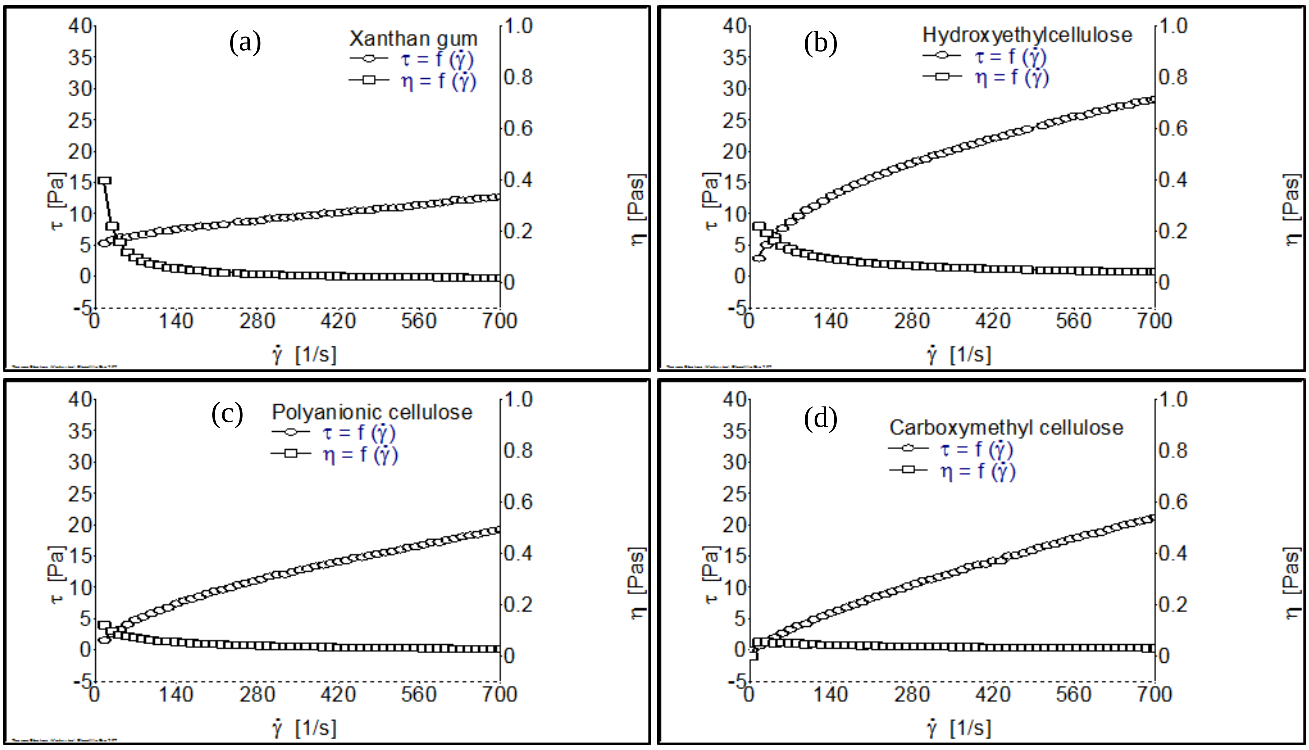

Figure 1. Shear stress (τ) and apparent viscosity (η) as a function of shear rate ($\dot{\gamma}$): (a) xanthan gum solution, (b) hydroxyethyl cellulose solution, (c) polyanionic cellulose solution and (d) carboxymethyl cellulose solution.

III. Results and discussion

Rheological behavior

The rheological curves of WSPs (XG, HEC, PAC and CMC HV) are shown in Figure 1. The shear stress and apparent viscosity as a function of shear rate of XG mixture at 0.4 wt. % is presented in Figure 1-a.

In contrast to the XG solution, the shear stress as a function of shear rate curve (Figure 1-b) displays the pseudoplastic behavior of the HEC-water system. This figure shows that the solution based on HEC exhibits pseudoplastic behavior. Therefore, experimental data were fitted to the model of Ostwald–De Waele Model [20]. In addition, the flow curve for the studied mass concentrations of CMC HV and PAC R clearly shows a non-Newtonian behavior without a yield stress (pseudoplastic behavior) as shown in in Figure 1-c and 1-d.

The flow curves of WSPs are exhibited in Figure 2. The H-B and Ostwald-de Waele models ($\tau_0 = 0$) were used to fit the shear rate dependence of the shear stress curves of WSPs solutions at 0.4 wt. % to verify the existence of the yield stress. The flow curves of PAC R, CMC HV and HEC show that these solutions behave as pseudoplastic fluids. A pseudoplastic fluid is a non-Newtonian fluid whose viscosity decreases with increasing shear rate. This behavior is characteristic of fluids without a yield stress. This property causes pseudoplastic fluids to exhibit lower viscosity at higher shear rates and higher viscosity at lower shear rates.

Table 2 presents the parameters as results of the fitting process by the H-B model of flow curves. In the case of the XG solution, the fitted parameter yield stress (τ_0) was 4.6 Pa. The flow curve of XG solution was found to be well fitted by H–B model (r = 0.999). On the contrary, the flow behavior of HEC, PAC R and CMC HV solutions did not indicate yield stress. Thus, flow curves were fitted by the Ostwald–de Waele equation.

With regard to the flow behavior index (n), the XG, HEC, PAC R and CMC HV solutions presented values of 0.623 $[Pa.s^n]$, 0.496 $[Pa.s^n]$, 0.589 $[Pa.s^n]$, and 0.780 $[Pa.s^n]$, respectively. It is obvious that WSPs show a pseudoplastic behavior with a flow behavior index less than 1 ($n < 1$). The pseudoplasticity of PAC R ($n = 0.589$) was greater than that of CMC HV ($n = 0.780$). Thus, CMC HV's chains are the easiest to orient under the effect of shearing compared to other HSPs (HEC and PAC R). McMullen et al. [20] confirmed that the CMC HV and HEC aqueous systems displayed shear-thinning behavior without yield stress.

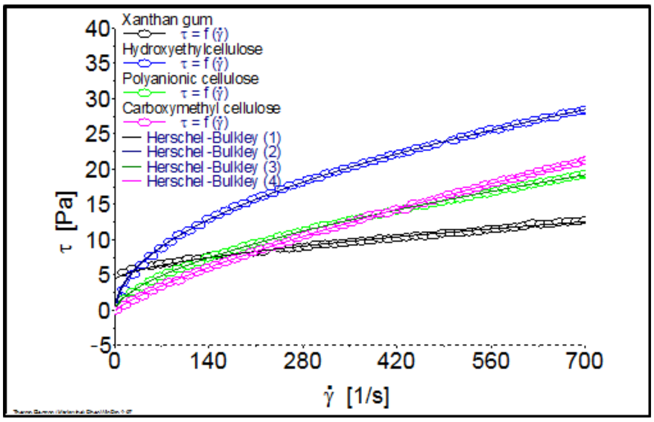

Figure 2. Shear stress (τ) of water-soluble polymers, the continuous solid lines correspond to the curve fitting to the H-B model.

Table 2. Rheological parameters values according to H-B and Ostwald-de Waele models.

water-soluble polymer	Rheological model	Parameters values
Xanthan gum	Herschel-Bulkley	$\boldsymbol{\tau_0 = 4.6}$ [Pa] K = 0.134 [Pa.s^n] n = 0.623
HEC	Ostwald-de Waele	$\tau_0 = 0$ K = 1.103 [Pa.s^n] n = 0.496
PAC R	Ostwald-de Waele	$\tau_0 = 0$ K = 0.405 [Pa.s^n] n = 0.589
CMC-HV	Ostwald-de Waele	$\tau_0 = 0$ K = 0.130 [Pa.s^n] n = 0.780

Edali et al [21] confirmed that pseudoplasticity is higher in both high molecular weight and DS (when the DS is 0.9 to 1.2). On the other hand, the HEC solution was more pseudoplastic compared to the CMC HV and PAC R solutions. The pronounced shear-thinning behavior of the HEC solution is attributed to its rigid chains.

The XG solution is known for its viscoplastic behavior. The existence of the yield stress (τ_0 = 4.6 Pa), as pointed out in the table 2 (XG solution), indicates that the XG solution is viscoplastic. Yield stress refers to the minimum shear force at which the polymer structure is destroyed, initiating flow. This behavior can reflect the strength of the network structure and is related to the network density of the XG [22].

the HEC solution showed the highest consistency index (k = 1.103 Pa·s^n) which relatively indicates high viscosity. The consistency index of XG, PAC R and CMC HV were 0.134, 0.405 and 0.130 Pa.s^n, respectively.

Apparent viscosity

This section discusses the relationship between apparent viscosity and shear rate, the Cross rheological model is used to provide a more comprehensive understanding of the rheological properties of WSPs. The apparent viscosity as a function of shear rate for polymer solutions are presented in Figure 3. The Cross model demonstrated a good fit with high correlation coefficient (r-value) to the observed apparent viscosity/ shear rate data of WSPs solutions.

Chalah et al. [16] reported that the Ostwald-de Waele and Cross models are in good agreement with experimental data for concentrations ranging from 0.7 to 1.7 % CMC-HV. The correlation coefficient value is greater than 0.999 for both the Ostwald-de Waele and Cross models. However, Salehi et al. [23] showed that the Ostwald–de Waele model is suitable to explain the rheological behavior of CMC HV solution. They indicated that the Power law equation demonstrated a good fit. On the other hand, Ouaer and Gareche [8] reported that the flow curves of HEC solutions are well fitted by the Cross model. The Cross model gave good ability to predict the flow behavior of HEC.

The apparent viscosities measured at low shear rate (10 1/s) were 0.526, 0. 0.268, 0.153 and 0.050 Pa.s for XG, HEC, PAC R and CMC HV, respectively. Subsequently, the apparent viscosities evaluated at a high shear rate (500 s^{-1}) were 0.026, 0.054, 0.029 and 0.025 Pa.s for XG, HEC, PAC R and CMC HV, respectively. Results showed that the apparent viscosities of XG and HEC solutions decreased rapidly with increasing shear rate. However, the phenomenon is less pronounced in the case of solutions based on PAC R and CMC HV. It is clear that the apparent viscosity of PAC R was significantly higher than that of CMC HV at low shear rates (> 100 1/s). Indeed, the functional properties of CMC HV depend on the degree of substitution of the cellulose structure, as well as the chain length of the cellulose backbone structure and the degree of clustering of the carboxymethyl substituents. From this point of view, the degree of substitution of PAC R is higher than that of CMC, which ensures better viscosity. Thus, PAC R macromolecules are generally considered to be stiffer than CMC HV macromolecules. In addition, the apparent viscosity of HEC solution was found to be significantly higher than PAC R and CMC-HV aqueous systems.

At high shear rates (210-700 1/s), XG, CMC HV, and PAC R all exhibit similar flow behavior where their flow curves are superimposed, indicating similar viscosity levels under these specific shear conditions. In contrast, Hydroxyethyl Cellulose (HEC) shows a higher flow curve, suggesting it maintains a higher viscosity compared to XG, CMC HV, and PAC R at the same high shear rates.

The rheological differences between WSPs determine their effectiveness and application in fluids. The anionic modified CMC HV and PAC R identified as pseudoplastic polymers, are used mainly in drilling muds in low-temperature drilling applications. Although PAC R is superior to CMC HV in filtration reduction, anti-salt, anti-collapse and high temperature resistance [24, 25]. PAC R have a higher and more uniform DS than CMC HV, resulting in better water solubility and more stable performance in solution. Economically, CMC HV and PAC R are generally more cost-effective than XG, known as an anionic natural polymer. However, XG offers superior thickening and stabilizing properties due to its viscoplastic behavior. From its neutral nature, HEC behavior is less affected by the salt content, which makes it stable in high salinity fluids. In addition, the choice of viscosifier depends strongly on the shear rate, especially at low shear rates. XG exhibits a very high viscosity at shear rates ranging from 0 to 40 1/s due to intermolecular forces like ionic interactions and macromolecular entanglement. While, thanks to the rigidity of its chains, the HEC solution showed high viscosity at low shear rate.

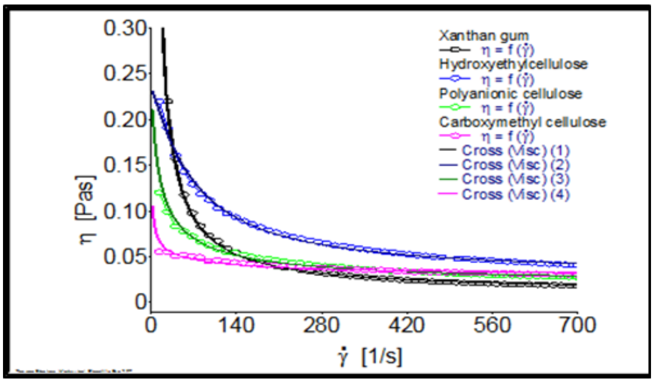

Figure 3. Apparent viscosity (η) of water-soluble polymers, the solid continuous lines correspond to the curve fitting.

IV. Conclusions

The rheological properties of four different water-soluble polymers were investigated. The experimental data were well fitted using the H-B and Ostwald-de Waele models. Knowledge of rheological properties such as rheological behavior, degree of shear-thinning, and viscosity is crucial for the selection and application of viscosifying polymers. The obtained results confirmed that:

1. The HEC, PAC R and CMC HV solutions showed pseudoplastic behavior at a concentration of 0.4 wt. %. However, even at a relatively low concentration (0.4 wt. %) XG solution behaved as a viscoplastic material, with a yield stress of 4.6 Pa.

2. The HEC solution (n = 0.496) is characterized by a higher degree of shear-thinning than PAC R (n = 0.589) and CMC HV (n = 0.780) solutions; the pseudoplasticity of HEC is highly pronounced.

3. At low shear rates, XG and HEC solutions showed higher apparent viscosity than CMC HV and PAC R solutions.

4. At high shear rates (200 to 700 1/s), the apparent viscosity of the WSPs is relatively low. Nevertheless, it should be noted that the HEC flow curve remains higher than the other curves (XG, CMC HV and PAC R).

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