

Performance Evaluation of the Rheological and Proppant Handling Property of Mucunaflagellipes as a Biopolymer in Hydraulic Fracturing Operations

Adali, F¹, Dosunmu, A² and Joel, O²

¹World Bank Africa Centre of Excellence for Oilfield Chemicals Research, University of Port Harcourt, Rivers State, Nigeria.

²Faculty of Engineering, Federal University, Otuoke, Bayelsa State, Nigeria.

*Corresponding author's email: franciseromosele@gmail.com

Abstract

Gear and its derivatives are the most common biopolymers used in hydraulic fracturing fluid design. This work is a step to sourcing alternative hydrocolloids for hydraulic fracturing fluid design. The rheological properties of Mucunaflagellipes, Cyamopsistetragonoloba (guar), and polyanionic cellulose-regular PAC-R were determined at temperatures of 27°C, 57°C, and 85°C respectively. It was found that the flow behaviour index increased with an increase in temperature and the type of polymers, with flow behaviour values of 0.1699 for Mucunaflagellipes at 27°C and 0.9453 for Cyamopsistetragonoloba at 85°C. The flow consistency index, however, decreased with an increase in temperature for the three polymers and there was no clear trend within the polymer. Cyamopsistetragonoloba exhibited the best rheology at higher shear rates while PAC-R exhibited the best rheology at higher temperatures and could be the best substitute for CT at higher temperatures. To analyze how the hydrocolloids handle proppants; the travel time of proppant in the fluid solution, the rheology of the fluid and the geometry of the proppant grains were used to compute the coefficient of drag, drag force and settling velocity of the polymer solutions. It was found that the PAC R gave the best drag force of $4.415 \times 10^{-5} \text{N}$ and Cyamopsistetragonoloba gave the best settling velocity of 0.8mm/s.

Keywords: Rheology, Galactomannan, Mucunaflagellipes, Cyamopsistetragonoloba, Polyanionic Cellulose-regular, Settling velocity

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1. Introduction

Hydraulic Fracturing is a stimulation technique used to improve the permeability of the rock and to create a network of pore spaces by pumping in fracturing fluids at high pressure and flow rate. The target is to improve the conductivity of the rock and the surface area contributing to flow. The success stories of hydraulic fracturing operations have made it economically feasible for the oil and gas industry to delve into the recovery of oil and gas from previously underdeveloped tight and ultra-tight reservoirs.

Fracturing fluids play a critical role in the hydraulic fracturing process for increasing the production of oil and gas in porous media such as sandstones, carbonates, and coal formations. Fracturing fluids must have sufficient viscosity to initiate and propagate hydraulic fractures and to suspend and transport propping agents deep into the created fracture (API RP 13M, R2018). Fracturing fluids should possess the following

rheological associated properties, sufficient viscosity, low treating pipe friction, shear stability, thermal stability, low to moderate fluid loss properties and controlled degradability. The key components in typical hydraulic fracturing fluid are the base fluid which could be water, oil or foam, viscosifiers or polymers, crosslinkers, breakers, and proppants. Some additives could be added based on the characteristic property of the fluid needed or the nature of the reservoir. Examples of such additives include biocides, buffers, clay stabilisers, fluid loss additives, friction reducers, surfactants etc.

Galactomannan is a heterogenous polysaccharide that is widely distributed in nature. They are mostly found in the endosperm of seeds in the Leguminosae family. They consist mainly of mannose and galactose in different ratios depending on the species. They can be used in their natural state or as derivatives. The properties of the galactomannan depend on their chemical

structures, chain length, availability of Cis-OH groups, and substitutes. An increase in substitution in the main chain leads to higher solubility. Galactomannan plays a critical role as an improving agent in processes where a hydrophilic system needs to be thickened, suspended, coated, etc. Galactomannan from the seeds of Leguminosae is a viable alternative source for polysaccharides used in the industries like guar and locus bean gums as they have similar sugar compositions. The variations in the degree of substitution and their crosslinking abilities may, however, lead to different chemical properties. Guar

(*Cyamopsistetragonoloba*), commonly known as guar, is a long-chain polymer with a high-molecular-weight in the order of 10^5 m. It is a leguminous galactomannan with mannose and galactose sugars in its chains which enhances its viscosifying property (Economides and Nolte, 2000). The mannose backbone of guar is insoluble in water, but the presence of the galactose branches enhances the solubility of the polymer. The ratio of mannose to galactose sugars may range from 1.6: 1, to 1.8: 1 (Economides and Nolte, 2000).

Due to occasional high prices and short supply of guar, there is a need for an alternative biopolymer to guar gum that can carry proppants, able to handle proppant packs, can work under high shear and temperatures, and can sustain a low viscosity at surface conditions but would have a higher viscosity downhole just before the fracturing job. Such a product is needed by the industry (Barati, 2014). Ma et al. (2018) stated that since guar gum and its derivatives cannot be used at elevated temperatures and oil well has gone deeper to HPHT zones, it is necessary to treat guar and its derivatives to be able to stand in high-temperature zones. They proposed the use of phosphates as a thermal stabilizer. They reported a 95% increase in thermal stability result for guar gum and an 80% increase for hydroxypropyl guar (HPG). They further stated that the thermal stability result is highest when the phosphate used is 1.84mmolL^{-1} of KH_2PO_4 for guar gum and 2.27mmolL^{-1} of K_2HPO_4 for HPG and the phosphate treatment demonstrates an excellent crosslinking ability. (Du et al., 2019) synthesized a hydraulic fluid combining an ionic polymer gel (hydroxypropyl trimethylammonium chloride guar-cationic guar, being crosslinked with a bola surfactant fluid (bola-carboxylate polypropylene glycol). They found that the hydraulic fluid showed improved properties and special characteristics due to the effects of the dual

systems. The viscosity of the fracturing fluid showed a sudden increase in value when the temperature increased and an excellent self-assembly recovery from shearing. They also showed that pH-responsive viscosity changed and there was low permeability impairment due to the formation of a network of structure and supramolecular microspheres at different pHs. Bo et al. (2018) opined that the most used synthetic polymer is polyacrylamide (PAM) and its derivatives. The PAM and its derivatives have polymers which are subjected to the hydrolysis of the acrylamide and this has a negative impact on the thermal stability of the fluid. Also, since these polymers lack cross-linkable groups, their viscosity, elasticity, and thermal stability are very limited. They proposed a novel non-residual fracturing fluid, which is produced by analyzing the structure of 2-acrylamido-2-methylpropanesulfonic acid (AMPS). Introducing the high-temperature-tolerant groups, 2-acrylamide and 2-methylenepropanesulfonic acid (AMPS), into the PAM, will improve the thermal tolerance of the fluid. Also, the acrylic acid, introduced into the polymer molecules provides carboxyl, which is capable of crosslinking with the multivalent transition metal ion and thus increases the viscosity of the fluid. Zhang (2019) proposed a novel fracturing fluid which would overcome the limitations of guar and its derivatives (high insoluble residue, poor shear resistance and pore throat plugging), the viscoelastic surfactants VES (loss of filtrates, difficulty in breaking gel and high cost of fluid) and the Hydrophobic associating water-soluble polymer HAWSP (high initial viscosity, equipment damage before operation). Prior to this work, some scholars proposed that sacrificing viscosity to yield a high elasticity far outweighs any advantage a high viscosity can render as high elasticity will improve sand carrying capacity and decreases friction between the fluid and the equipment. The elasticity of polymer solutions can be improved by using hydrophobic association interactions, electrostatic bridge effects, hydrogen bonds and Van Der Waal forces. High Elasticity, Low Viscosity (HELV) was designed by copolymerizing acrylamide (AM), acrylic acid (AA), 4-isopropenylcarbamoylbenzene sulfonic acid (AMBS) and N-(3-methacrylamidopropyl)-N, N-dimethyldodecan-1-aminium (DM-12). A copolymer solution with low viscosity and high elasticity was synthesized by introducing a benzene ring, sulfonates, and long

hydrophobic chains into the polymer structure. Excellent viscoelasticity, and thixotropy novel hydraulic fracturing fluid were produced. Zhao et al. (2019) proposed a new type of fluid with two kinds of liquid injected simultaneously and with no proppant used. The two types of fluid act as fracturing fluid and supporting solids. One kind of fluid called phase change liquid (PCL) will convert to solid at high temperature and serve as a proppant to resist fracture closure. The other non-phase change liquid (NPCL) has no phase change in the whole process and acts like the conventional fracturing fluid. Antosik (2017) did a study on the influence of hydroxyl groups and oxygen atoms with chain length and branching of carrier fluid on the rheological and electrokinetic properties of shear thickening time (STF). Ethylene glycol, triethylene glycol, 1, 3-propanediol, glycerine, poly (propylene glycol) of different molecular weights and poly (propylene glycol) triol were used as carrier fluids (dispersants). Silica powder with an average particle size of 100nm was used as the solid phase. Zeta potential, particle size distribution, steady state and dynamic rheological measurements were conducted. The results showed that a different amount of -OH group and oxygen atoms together with chain length and branching of carrier fluids have a significant effect on the intermolecular interactions and that it is possible to control the rheological properties of a hydraulic fracturing fluid. Geri et al. (2019) proposed the use of high viscosity friction reducers (HVFRs), which typically are high molecular weight polyacrylamides, as a substitute for biopolymers. The fluid was used in over 26 case studies, and it showed superior proppant transport capability, almost 100% retained conductivity, cost reduction, minimizing chemicals usage by 50%, less operating equipment on location, reduced water consumption by 30%, and fewer environmental concerns. PAC R is a member of the polyacrylamides group and together with guar is the control fluid in this work. Cao et al. (2021) investigated a novel synthetic polymer, in the polyacrylamide family. Polyacrylamide-co-acrylic acid-co-2-acrylamido-2-methyl-1-propanesulfonic acid (P3A) for its use as a tertiary oil recovery polymer in high temperature and high salt reservoirs. They reported a better result than guar at higher temperatures and high salt reservoirs. They concluded that polyacrylamide is a good substitute for the thermally unstable guar at higher temperatures and high salt reservoirs.

Mucunaflagellipes is of the Leguminosae family and sub-family Papilionacea. It is native to West Africa. It is high-yielding and could be cultivated more than once a year. It bears pods that contain 3-4 seeds per pod. The seeds are normally dark brown to black and could sometimes be speckled. Nwokocha (2009) subjected different concentrations of MF gum powder (0.1%, 0.2%, 0.5%, 1.0%, 2.0% and 3.0%) in distilled water and left it overnight at ambient temperature. It was deduced from the work that the endosperm of MF is made of 46.73-74.76% of the whole seed with a mean of 67.15%. The defatted endosperm yielded $32.6 \pm 1.97\%$ of gum. He applied the Mark-Houwink equation which gave an average molecular mass of $2.1 \times 10^6 \text{ gmol}^{-1}$ which is similar to the value reported for galactomannans like carob gum and guar gum. Nwokocha (2012) characterized *Mucunaflagellipes*, *Detariummicrocarpum*, and some other possible biopolymers native to the West African climate. He opined that with the molecular weight, the endosperm yield, and the gum yield of the MF, it is a very good candidate as a viscosifier in industrial processes.

2. Materials and methods

2.1 Experimental procedure

The MF seed was bought at Isiokpo Bush Market, while the CT and PR were bought from Joechem Chemicals, Choba, Port Harcourt. The *Mucunaflagellipes* seed is dehulled as in Fig. 1 and the resulting endosperm is pulverized using a hammer mill as in Fig. 2. The pulverized seed is blended with a blender to have a fine powder. The powder is wrapped in a crystalline filter paper and placed in a Soxhlet extractor to extract the oil in it. This was done using propanol. The content was left in the Soxhlet extractor for three days in a continuous reflux process. The resulting powder was placed in an oven at a temperature of 80°C for 6 hours. The resulting MF powder together with the already process CT and PR were mixed individually at a concentration of 10g per litre of water. The three samples were placed in a Fann viscometer, and the dial readings are taken at RPM of 3, 6, 100, 200, 300, and 600 RPM and 27°C. The procedure was repeated at temperatures of 57°C and 85°C respectively. With MF, CT, and PR at a concentration of 10g/l in 3 burettes, the travel time of proppant sand particles of 78% 0.002m grain diameter (ASTM 10) down the burettes was taken

and used as the raw data to compute the drag force and the settling velocity.



Fig. 1: *Mucunaflagellipes* seed



Fig. 2: Pulverized *Mucunaflagellipes*

2.2 Constitutive equations development

The fluids exhibit Herschel-Buckley behaviour. The following constitutive equations were used for the rheology, drag velocity and settling velocity (Bourgoyne et al., 1986).

$$\tau = \tau_y + K\gamma^n \quad (1)$$

$$P_v = \theta_{600} - \theta_{300} \quad (2)$$

$$\tau_y = \theta_{300} - P_v \quad (3)$$

$$\tau = 0.01066 \times \theta_i \times N \quad (4)$$

$$\gamma = 1.703 \times RPM \quad (5)$$

$$n = 3.32 \log \log \frac{\theta_{600}}{\theta_{300}} \quad (6)$$

$$k = \frac{\theta_{300}}{511^n} = \frac{\theta_{600}}{1022^n} \quad (7)$$

$$C_D = \frac{F_D}{\frac{1}{2}(\rho v^2)(\pi R^2)} \quad (8)$$

$$C_D = \frac{4gd}{3v^2} \left(\frac{\rho_s - \rho}{\rho} \right) \quad (9)$$

$$R_e = \frac{\rho v^{2-n} d^n}{m} \quad (10)$$

$$B_i = \frac{\tau_o}{m \left(\frac{v}{d} \right)} \quad (11)$$

Equations (9), (10) and (11) can be solved using different values of the slip velocity (v) by trial and error. The result can be validated using semi-empirical correlation for drag on spheres in Herschel-Bulkley model liquids (Atapattu et al., 1995).

$$C_D = \frac{24}{R_e} (1 + B_i) \quad (12)$$

$$F_D = \frac{\pi d^3 (\rho_s - \rho) g}{6} \quad (13)$$

For,

$$10^{-5} \leq Re \leq 0.36$$

$$0.25 \leq Bi \leq 280$$

$$0.13 \leq n \leq 0.95$$

The values of B_i , R_e , the diameter of proppant grains, the density of the proppant grains and the density of the carrier fluids are inputted into Equations (12) and (13) to compute the coefficient of drag and the drag force.

3. Results and discussion

The rheology of *Mucunaflagellipes* (MF), *Cyamopsistetragonoloba* (CT), and Polyanionic-cellulose filtration control additive PAC R (PR) were taken at 27°C, 57°C, and 85°C, after 10 sec and after 10 min. The converted readings using equations (4) and (5) are highlighted in Tables 1, 2 and 3. The sizes from sieve analysis are run on the sieve conversion chart in appendix 1 for the diameter of the proppant sand. Table 1 shows the dial reading of MF at temperatures 27°C, 57°C and 85°C, using Equations (1) to (7). The shear strain and the shear stress at 27°C, 57°C and 85°C are computed and tabulated in Table 1. The shear stress is observed to decrease with an increase in temperature for *Mucunaflagellipes*. Also, there is a general decrease in shear stress with a decrease in shear strain. The flow behaviour index, n and the consistency factor, k computed from Equations (6) and (7), decrease with an increase in temperature.

Table 1: Rheology of *Mucunaflagellipes* at 27°C, 57°C and 85°C

Shear strain (S ⁻¹)	Stress (lbf/100ft ²) 27°C	Stress (lbf/100ft ²) 57°C	Stress (lbf/100ft ²) 85°C
1021.80	0.0959	0.0746	0.0640
510.90	0.0853	0.0640	0.0426
340.60	0.0533	0.0533	0.0320
170.30	0.0426	0.0426	0.0320
10.22	0.0320	0.0213	0.0213
5.11	0.0320	0.0213	0.0213
n	0.1699	0.2223	0.5848
k	0.0295	0.0160	0.0011

Table 2 shows the dial reading of CT at temperatures 27°C, 57°C and 85°C, using equations 1 to 7. The shear strain and the shear stress at 27°C, 57°C and 85°C are computed and the results are presented in Table 2. A general decrease in shear stress is observed with increase in temperature for *Cyamopsis tetragonoloba*. Also, there is a general decrease in shear stress with a decrease in shear strain. However, the flow behaviour index of *Cyamopsis tetragonoloba* increases with an increase in temperature. The

consistency factor decreases with an increase in temperature.

Table 3 shows the dial reading of PAC R at temperatures 27°C, 57°C and 85°C, using equations 1 to 7 to generate the table from the raw data on appendix 2, Table 4. The shear strain and the shear stress at 27°C, 57°C and 85°C are computed and the results are presented in Table 3. A general decrease in shear stress is observed with increase in temperature for PAC R. Also, there is a general decrease in shear stress with a decrease in shear

strain. The flow behaviour index has a little twist; it goes higher at 57°C and goes slightly lower at 85°C. The consistency factor however, follows the regular pattern of decreasing with increase in temperature.

Table 2: Rheology of *Cyamopsistetragonoloba* at 27°C, 57°C and 85°C

Shear strain (S ⁻¹)	Stress (lbf/100ft ²) 27°C	Stress (lbf/100ft ²) 57°C	Stress (lbf/100ft ²) 85°C
1021.80	0.75686	0.68224	0.55432
510.90	0.54366	0.3731	0.28782
340.60	0.44772	0.29848	0.17056
170.30	0.3198	0.17056	0.11726
10.22	0.06396	0.04264	0.03198
5.11	0.0533	0.03198	0.02132
n	0.4772	0.8705	0.9453
k	0.0277	0.0016	0.0008

Table 3: Rheology of PAC R at 27°C, 57°C and 85°C

Shear strain (S ⁻¹)	Stress (lbf/100ft ²) 27°C	Stress (lbf/100ft ²) 57°C	Stress (lbf/100ft ²) 85°C
1021.80	0.46904	0.44772	0.40508
510.90	0.28782	0.24518	0.22386
340.60	0.2132	0.20254	0.18122
170.30	0.12792	0.1066	0.09594
10.22	0.03198	0.03198	0.02132
5.11	0.03198	0.03198	0.02132
n	0.7043	0.8685	0.8554
k	0.0036	0.0011	0.0011

Fig. 3 shows the rheology of MF at different temperatures and 10g/l concentration. From Fig. 3, the MF showed better rheology at 27°C, that is, at laboratory temperature. However, at higher temperatures of 57°C and 85°C, there is a marked reduction in rheology. The interesting part of this finding is that the curves are closely knitted. It is an indication that there is no marked difference in rheology at 57°C and 85°C. Also, the yield point is highest at 27°C with a yield point of 0.0336 lbf/100ft². The yield points at 57°C and 85°C are 0.0214 lbf/100ft² and 0.0264lbf/100ft² respectively.

The shear stress-shear strain plot as in Fig. 4 showed the rheological behaviour at 27°C. It is observed that there is equidistance among the three temperatures. This is an indication of the steady denaturing of the polymer with an increase in temperature. It is an indication that there is no marked difference in rheology at 57°C and 85°C. Also, the yield point is highest at 27°C with a yield

point of 0.1293 lbf/100ft². The yield points at 57°C and 85°C are 0.0495 lbf/100ft² and 0.0191lbf/100ft² respectively. The plot of the shear-stress against shear-strain for the PR at different temperatures as is in fig 5 showed a close knitted behaviour at lower shear strain and an equidistance close-knitted behaviour at higher shear strains. It is an indication that there is no marked difference in rheology at 57°C and 85°C. Also, the yield point is highest at 27°C with a yield point of 0.0449lbf/100ft². The yield points at 57°C and 85°C are 0.0371lbf/100ft² and 0.0284lbf/100ft² respectively.

Fig. 6 shows the three biopolymers at 27°C. CT shows the best rheology followed by PR and then MF. There is also an obvious gap in the rheology of the three biopolymers. This implies that there is a clear difference in their performances in Laboratory conditions, CT > PR >MF. Recall that CT is the industry favourite at lower temperatures.

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Also, CT has the best yield point at 0.1293lb/100ft².

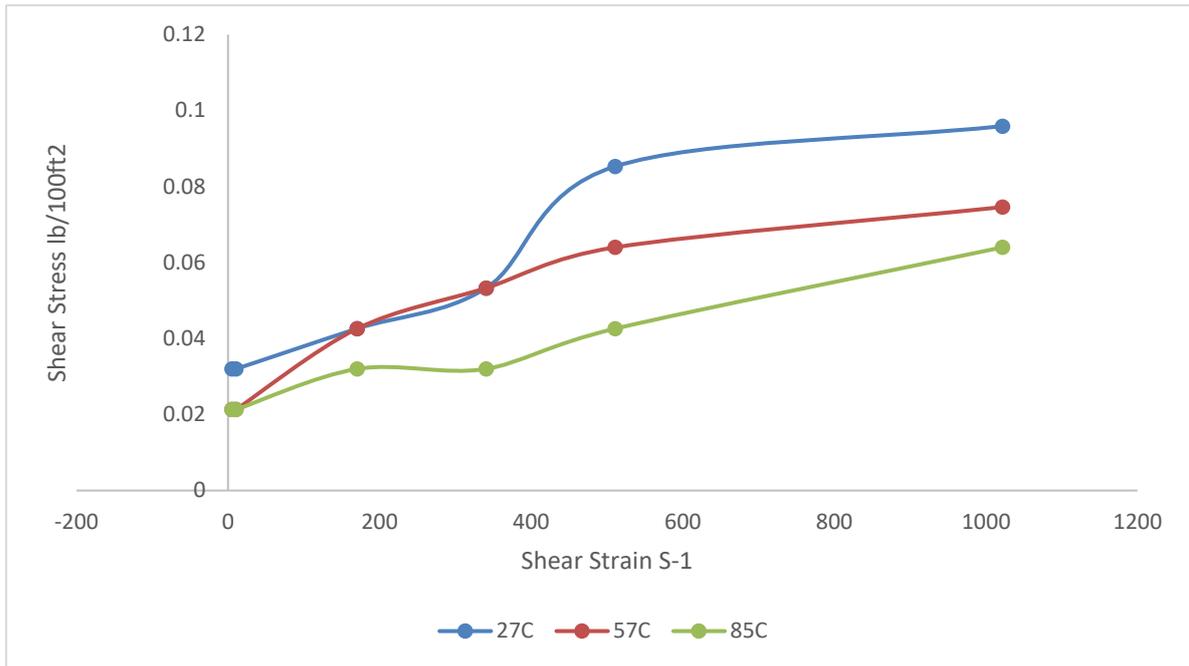


Fig. 3: Rheology of MF at different temperatures and 10g/l concentration

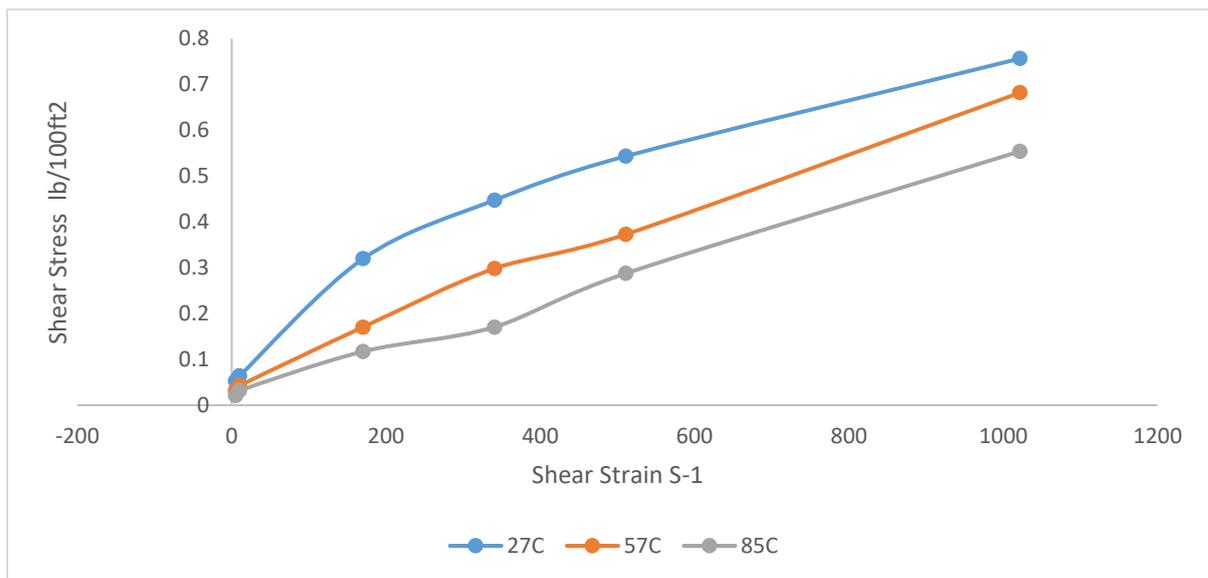


Fig. 4: Rheology of CT at different temperatures and 10g/l concentration

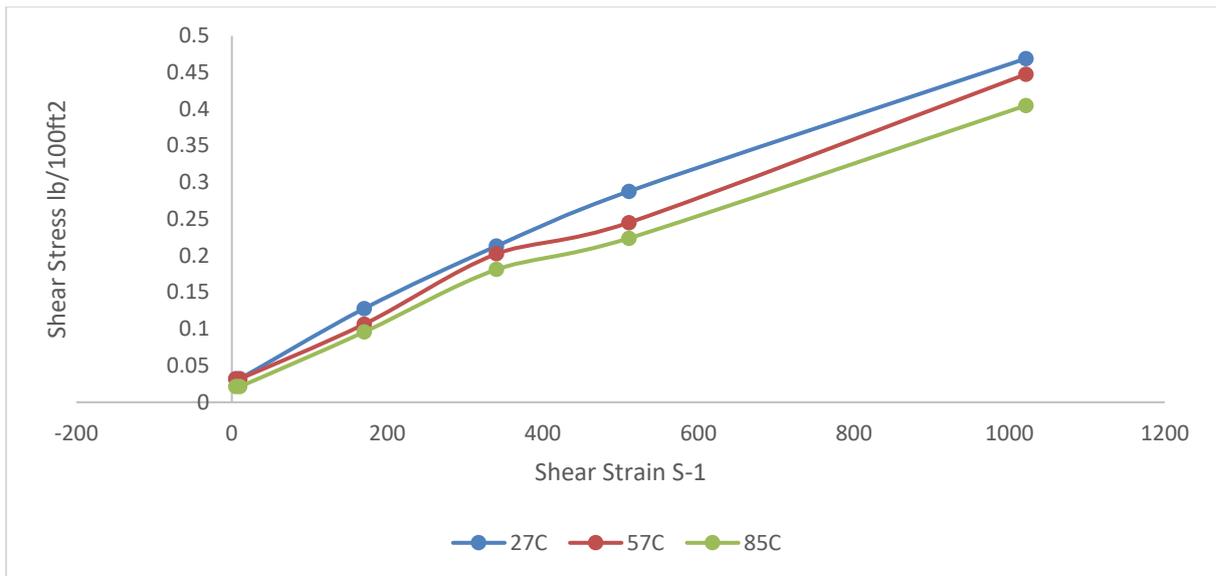


Fig. 5: Rheology of PR at different temperatures and concentration of 10g/l

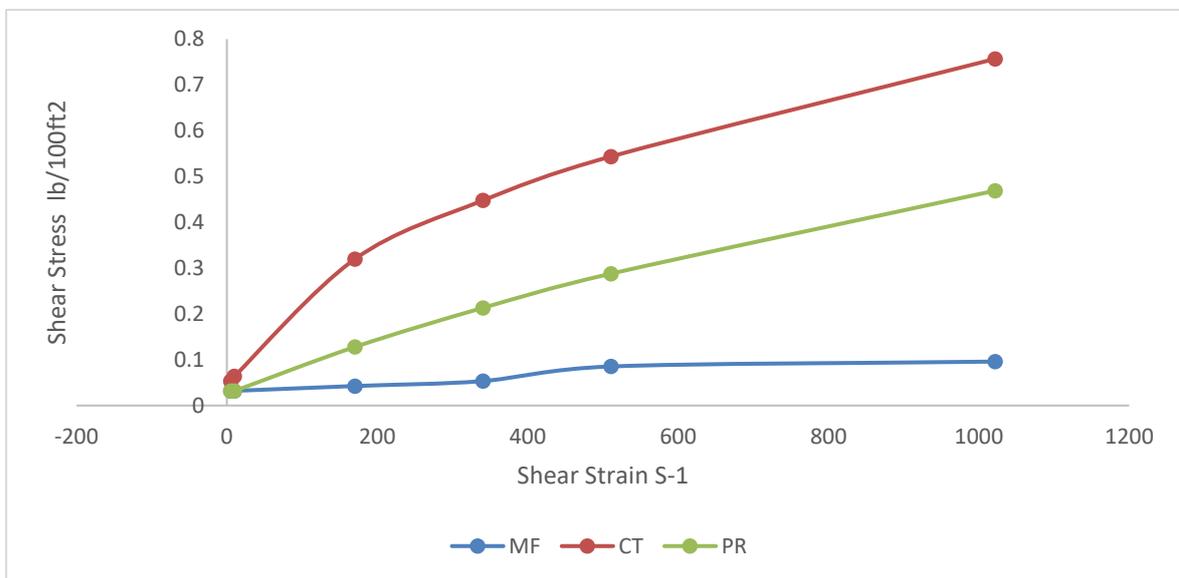


Fig. 6: Rheology of MF, CT, and PR at 27°C and 10g/l concentration

The Fig. 7 shows the three biopolymers at 57°C. CT shows the best rheology followed by PR and then MF. There is also an obvious gap in the rheology of the three biopolymers. It is observed that the gap between the MF and PR remains as huge as in 27°C whereas that of CT and PR closes gradually at 57°C, to show an improvement of PR at a higher temperature.

For Fig. 6, the rheological behaviour of the three polymers at a laboratory temperature of 27°C showed CT as the best with a wide margin. The PR is the second best. However, at a higher temperature of 57°C for fig 7, the gap between CT and PR has been narrowed with MF still languishing at the bottom. And at 85°C for Fig. 8, there is a close-knit in the properties of CT and PR,

at lower shear rates and close enough properties at higher shear rates. The MF still languished at the bottom. This is an indication that the PR could be a good substitute for CT at a higher temperature hydraulic fracturing job.

Fig. 9 shows a steady decline in the consistency index, k of MF and CT. The consistency index of PR, however, remains the same throughout the temperature range. This further shows the efficacy of PR at higher temperatures. MF still has the temperature denaturing problem that is synonymous with CT.

Table 4 shows the proppant handling capacity of the three polymers. The table was generated using equations (8) to (13). PR shows the best drag

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or buoyancy force of $4.415 \times 10^{-5} \text{N}$. This implies that PR has the least tendency for the polymer sediment to settle out of the solution. The settling

or terminal velocity is best with CT, which has the least settling velocity of 0.8mm/s and will thereby take the least time to settle out of its solution.

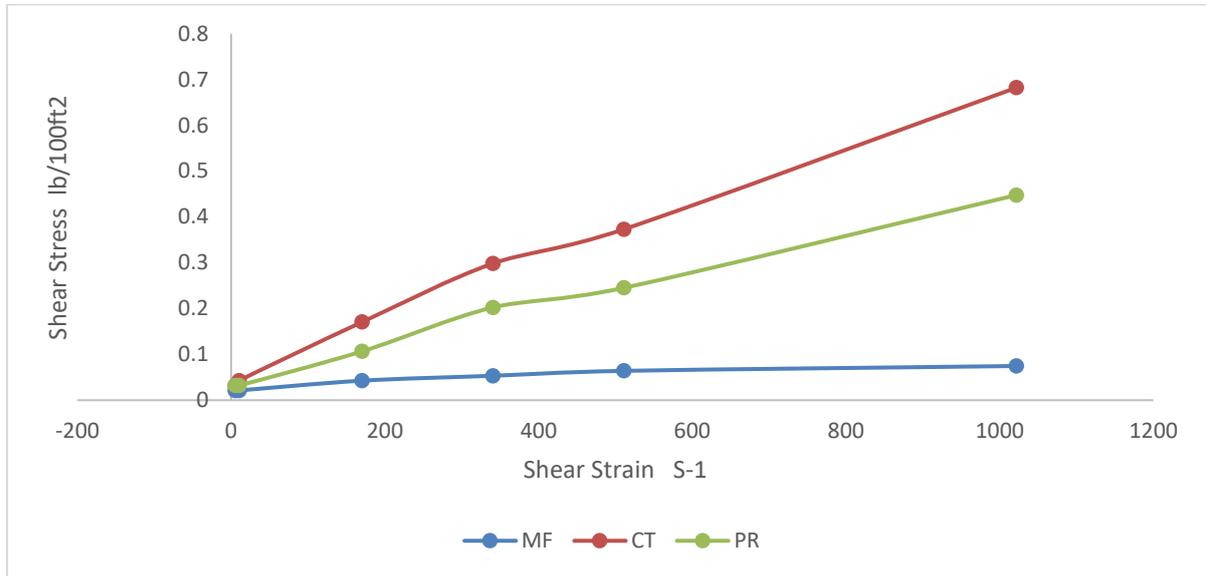


Fig. 7: Rheology of MF, CT, and PR at 57°C and 10g/l concentration

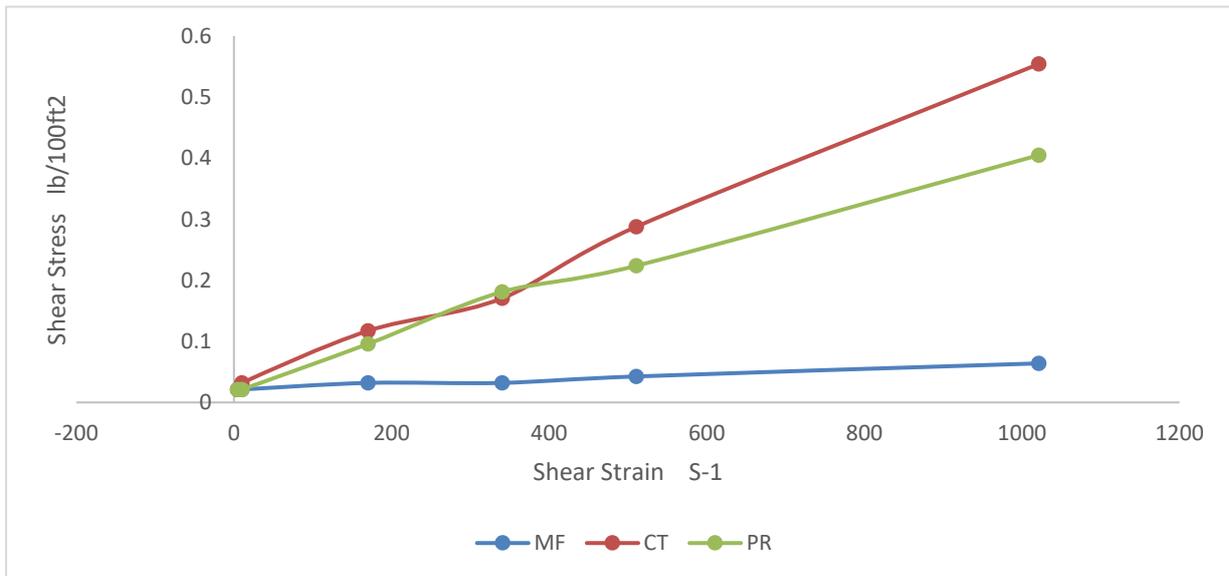


Fig. 8: Rheology of MF, CT, and PR at 85°C and 10g/l concentration

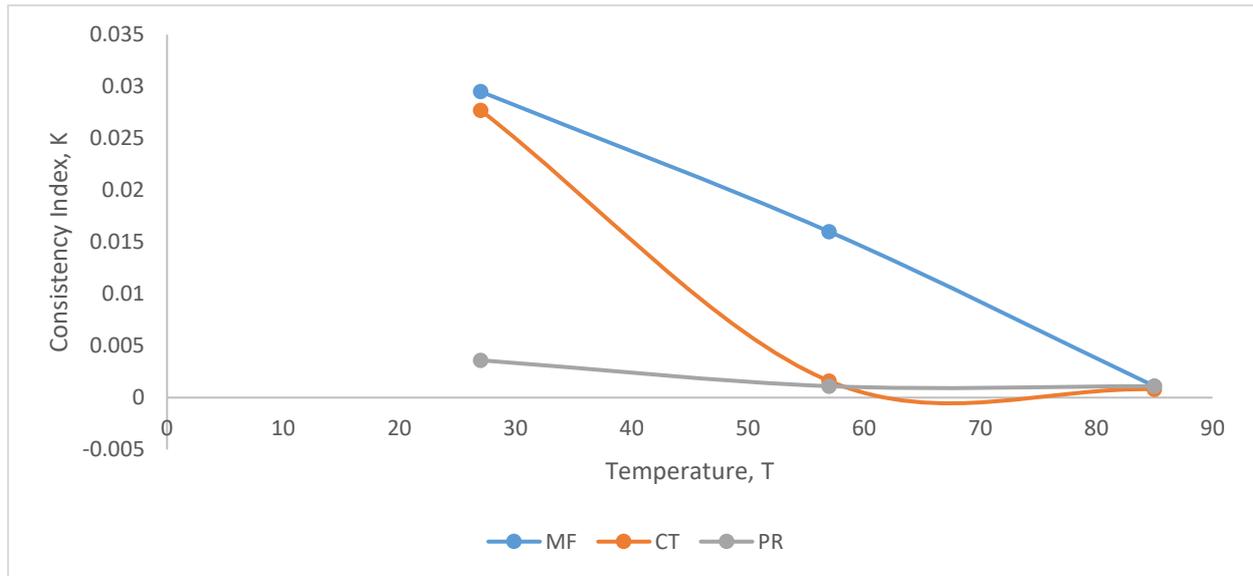


Fig. 9: Consistency index (k) of the three polymers against temperature

Table 4: Summary of results from the drag force and settling velocity experiment

	Reynold number, Re	Bingham number, Bi	Drag coefficient, CD	The drag force, F_D (N) $\times 10^{-5}$	Settling velocity, v (mm/s)
MF	64	261	6,879.13	1.617	2.2
CT	3.05	264.8	40,679	4.205	0.8
PR	72	220	5365.86	4.415	2.4

4. Conclusion

Based on the results obtained from the study, the following conclusions can be drawn:

- The carrier fluid designed from the three polymers exhibited the Hershel Bulkley fluid behaviours.
- The flow behaviour index was determined for the three polymers, and it was deduced that the flow behaviour index increased with an increase in temperature.
- The values for the commercial CT were highest, followed by PR and then for MF at a concentration loading of 10g/l.
- The flow consistency index, however, decreased with an increase in temperature for the three polymers and there is no clear trend within the polymer.
- CT exhibited the best rheology at higher shear rates and is still the best polymer at higher shear rates.
- The PR exhibited the best rheology at higher temperatures and could be the best substitute for CT at higher temperatures.
- PR has the best drag force and CT has the best settling velocity.

- The MF is not a suitable substitute for the commercial CT.

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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