

Original Research Article

Comparative Analysis of the Effects of Monovalent and Divalent Ions on Imported Biopolymer-Xanthan Gum and Locally Formulated Biopolymers-Gum Arabic and Terminalia Mantaly

ABSTRACT

Aim: Polymer flooding is used for enhanced oil recovery. Only polymers that can withstand harsh environments work best. HPAM is mostly the polymer used for enhanced oil recovery because it is available and cheap, but it does not withstand high temperatures and high salinity reservoirs. Xanthan Gum withstands high temperatures and high salinity reservoirs, but it is expensive and plugs the reservoir. The aim of this study is to compare the salinity stability of gum Arabic and Terminalia Mantaly, a novel biopolymer, with commercial Xanthan gum.

Study Design: Locally formulated biopolymers from gum Arabic exudates bought from Bauchi State in Nigeria and from Terminalia Mantaly exudates obtained from the University of Port Harcourt. The appropriate rheological tests were carried out at the laboratory.

Place and Duration of Study: The laboratory experiments were carried out at the department of Petroleum Engineering, Covenant University, Ota in Ogun State of Nigeria between 2020 and 2021.

Methodology: The gum Arabic, Terminalia Mantaly and Xanthan Gum powders were dissolved in deionized water to get various concentrations in ppm. The polymers were mixed and kept for 24 hours to achieve a homogenous solution. The Automated OFITE® Viscometer at different revolutions per minute (RPM) of 3 (Gel), 6, 30, 60, 100, 200, 300, and 600 was used to measure the rheological properties of the various concentrations before Sodium Chloride (NaCl) and Calcium Chloride (CaCl₂) of various concentrations were added and allowed to hydrate for another 24 hours before measuring their rheological properties again.

Results: The study showed that Xanthan Gum, Gum Arabic, and Terminalia Mantaly biopolymers can be used in high salinity reservoirs. Terminalia Mantaly, a novel biopolymer, is insensitive to salinity in monovalent and divalent ions.

Conclusion: Xanthan gum exhibited high viscosity even at low concentrations. Gum Arabic exhibited good tolerance to salinity at NaCl 3.5%. Terminalia Mantaly was very stable with both monovalent and divalent ions. Divalent ions have more effects on polymers than monovalent ions in reservoirs.

Recommendation: It is recommended that Terminalia Mantaly be investigated more, as it can replace imported biopolymers for Enhanced Oil Recovery (EOR).

Keywords: Gum Arabic; Monovalent and Divalent Ions; Polymer Solutions Rheology; Stability; Terminalia Mantaly; Xanthan Gum.

1. INTRODUCTION

1.1 Background

Polymer flooding is a promising Chemical Enhanced Oil Recovery (cEOR) method used to recover residual oil, especially in heavy oil where waterflooding is not efficient due to viscous fingering (1-6). In the oil and gas industry, two types of polymers are mostly used: synthetic polymers like hydrolyzed polyacrylamide (HPAM) and its derivatives, and biologically produced biopolymers like Xanthan Gum (XG) and cellulose (7–10). HPAM has been used for the majority of the field polymer flooding because it is cheap and available, while biopolymers like xanthan gum have been used in very few fields because of their high cost and plugging abilities (11-14). Hydroxyethyl cellulose (HEC) is used for high salinity, low temperature (HSLT) reservoirs due to their tolerance to high salinities where precipitation occurs in HPAM (15).

Although HPAM is cheap and available, it has issues in high temperature and high salinity (HTHS) reservoirs and suffers from polymer degradation. Xanthan gum, on the other hand, withstands high temperatures and high salinity reservoirs, but it is expensive and easily degrades when bacteria are present in the reservoirs (16-19). There are HPAM derivatives that have been synthesized from HPAM to overcome high salinity or high temperature, like Hydrophobically Associated Polyacrylamides (HAPAM) and Comb Shaped Modified HPAM-A High Salinity Tolerant Polymer (KYPAM), but they are still synthetic in nature, non-biodegradable and will be harmful to the environment (8, 20-22).

1.2. Xanthan Gum

Xanthan gum is a non-ionic bacterial polysaccharide produced by the fermentation of a cellulosic backbone consisting of five monosaccharides by the bacterium *Xanthomonas campestris* to give a pentasaccharide repeating unit. Xanthan gum is a biopolymer. It is used in the pharmaceutical industries, agricultural sectors, food industries, cosmetics, textiles, paints, and the petroleum industries. The chemical structure of Xanthan gum reveals that it has a rigid structure that can withstand HTHS, mechanical shear and divalent ion concentration (6, 23-24).

Xanthan gum has a high molecular weight ($>2 \times 10^6$ g/mol), and its solutions exhibit shear thinning, higher viscosity, ease of penetration into low permeability zones, and drag reducing criteria in subsurface environments (25-27). Figure 1 presents the chemical structure of xanthan gum, showing a single glucuronic acid unit, two mannose units, and two glucose units of molar ratios of 2.0, 2.0, and 2.8, respectively (7).

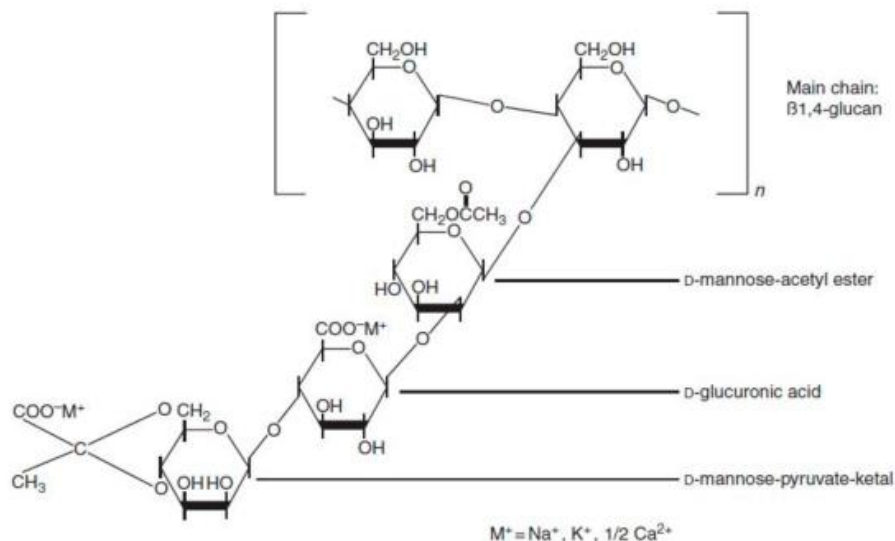


Fig. 1. Chemical Structure of XG showing both D-Mannose and D-glucuronic acid units linked to the backbone of the glucose, while (M⁺) is the cation binding sites (Source, Muhammed et al, 2020(?)).

1.3. Gum Arabic (GA)

Gum Arabic is the name given to a natural plant polysaccharide gum exudates of hardened sap from *Acacia Senegal* (*Senegalia Senegal*) and *Acacia Seyal* (*Vachellia*), deciduous trees. Gum Arabic belongs to a family of trees called Leguminosae. Gum Arabic is the oldest plant polysaccharide and it has other names like Acacia Gum, Arabic Gum, Indian Gum, Gum Acacia, Acacia, Senegal Gum, Gum Hashab, Gum Talha, Gum Sudani etc. It is an important cash crop for most countries in sub-Saharan Africa, like Sudan, Nigeria, Mauritania, Mali and Senegal. Sudan is the world's leading producer of gum Arabic, which has greatly benefited the country's economy. Gum Arabic is highly soluble in water, with good emulsifying properties but much lower viscosity compared to other biopolymers like xanthan gum (28-30). Gum Arabic is primarily used as a natural emulsifier. Other industries like pharmaceuticals, ceramics, printing, textiles, inks, paper, adhesives, cosmetics, paint, glue, chewing gum, photosensitive chemicals, and pyrotechnics make use of gum Arabic (31-32).

Gum Arabic's trade in the middle age was controlled by the Turkish Empire and it was nicknamed Turkey Gum (33). Gum Arabic was transported to Europe from Arab countries via Arabic ports and it gave rise to the name gum Arabic (34).

GA is a complex mixture of polysaccharides and glycoproteins (GP's) with a pH ranging from near neutral to mild acidity. It exists as a blended calcium, magnesium, and potassium salt of a polysaccharide acid known as Arabic acid. The framework of GA is made up of 1,3-connected-d-galactopyranosyl units. The side chains are made up of two to five 1,3-connected-d-galactopyranosyl units, joined to the primary chain by 1,6-linkages. Both the fundamental and the side chains contain units of α-l-arabinofuranosyl, α-l-rhamnopyranosyl, β-d-glucopyranosyl, and 4-O-methyl-β-d-glucopyranosyl, the last two generally as end units.

The chemical composition of GA can change with weather conditions, soil conditions, and the age of the tree and location of the tree (30, 35).

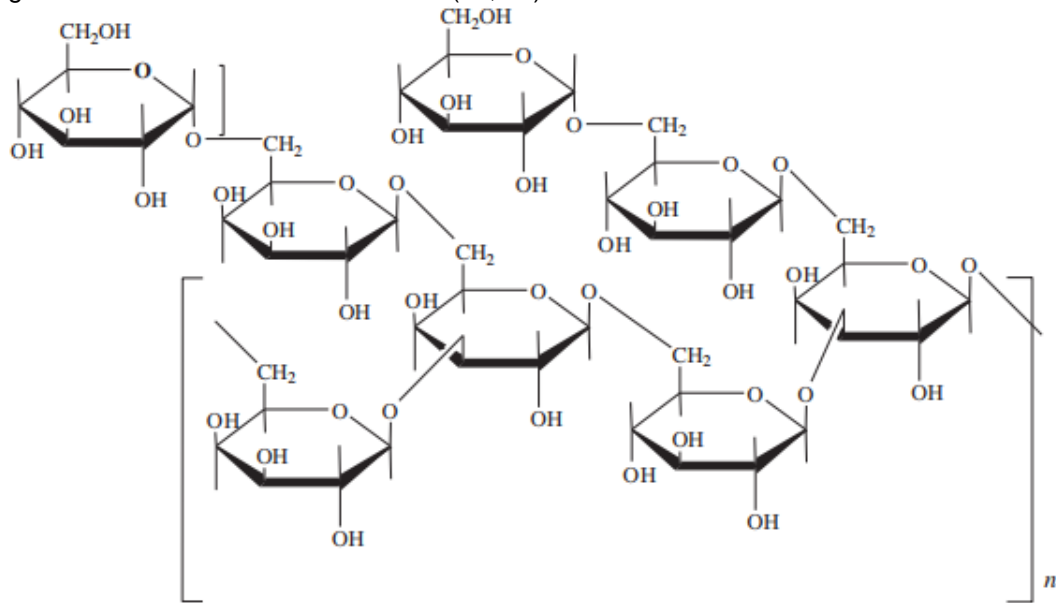


Fig. 2, presents the chemical structure of gum Arabic, showing L-arabinose, L-rhamnose and D-glucuronic acid and 1, 3-linked β-D-galactopyranosyl units (Source, Dave et al, 2018 (?)).

1.4. Terminalia Mantaly (TM)

Terminalia Mantaly is an evergreen or deciduous tree that grows up to 10–20 m with an erect stem and neat, conspicuously layered branches. Its bark and branches have been used for medicinal purposes. The bark and wood of Terminalia Mantaly contain tannins and are used as dyes, inks, tattoos, stains, and mordants. Terminalia Mantaly belongs to a family of trees called Combretaceae (36-37).

The effect of Terminalia Mantaly leaves in water-based mud as an additive to improve drilling mud properties has been investigated (38). The rheological properties of Terminalia Mantaly exudate have been investigated as a drilling mud additive. The study showed that Terminalia Mantaly polymer has good alkaline at pH above 7 and stable rheological properties as both salinity and temperature are increased. This is as a result of the stable repulsive charges polyelectrolyte of the polymer when salinity increases (39).

For this research, the focus is on the comparative effects of monovalent and divalent ions on Xanthan gum an imported biopolymer with gum Arabic and Terminalia Mantaly locally sourced biopolymers, to determine if either GA or TM can withstand the action of monovalent and divalent ions, since both GA and TM are cheap, available and ecofriendly.

Table 1. List of Physicochemical properties of Terminalia Mantaly gum (Source, Odeniyi et al, 2017(?))

S/No	Parameters	Values
1.	Particle Diameter (μm)	263.10
2.	Angle of Repose ($^{\circ}$)	57.80
3.	Particle Density (g/cm^3)	1.32
4.	Bulk Density (g/cm^3)	0.090
5.	Tapped Density (g/cm^3)	0.139
6.	Hausner's Ratio	1.54
7.	Carr's Index	35
8.	Swelling Index	8.4
9.	Water Absorption Capacity	10.71
10.	pH	7.31

Based on the comparison of native and microwaved TM at 20s and 60s, Table 1 was developed to show the list of physicochemical properties of Terminalia Mantaly gum (40). This has helped to understand the physicochemical properties of TM and its application.

2. MATERIAL AND METHODS

2.1 Materials.

For the project, the following materials were used,

Table 2 List of Materials

S/N	List of Materials	Description
1.	Gum Arabic (GA)	Purchased from Bauchi State and processed.
2.	Terminalia Mantaly (TM)	Collected from the University of Port Harcourt and processed,
3.	Xanthan Gum (XG)	Commercial biopolymer imported.
4.	Sodium Chloride (NaCl)	Monovalent Ion for brine preparation
5.	Calcium Chloride (CaCl_2)	Divalent Ion for brine Preparation

Table 2 shows a list of the materials used for this project. The XG used was an imported biopolymer, and it was used without any modification or purification. The TM exudates were collected from a TM tree located at the University of Port Harcourt. The TM exudates were processed using the method used by Odeniyi et al., 2017 (40). The GA exudates were purchased from a local market in Bauchi State and sent down to Port Harcourt in a sealed container. It was processed in Port Harcourt.

2.2 Preparation of Gum Arabic Powder

The gum Arabic was purchased from Bauchi State. The gum Arabic exudates were hand-picked to remove the impurities, and the clean exudates were pulverized with a sledgehammer in a clean, strong sack bag and blended afterwards. The GA powder was sieved with a 250 μm sieve initially and then with a 125 μm sieve to obtain the desired gum Arabic powder size.

2.3 Preparation of Terminalia Mantaly Powder

The gum exudates were collected from TM trees located at the University of Port Harcourt and dried in an oven at 50 °C for 24 hours. The gum exudates were pulverized with a blender to get TM powder. The powdered gum was hydrated in a mixture of double-strength chloroform (CHCl₃) and water for five days while being stirred intermittently. Extraneous materials and some undissolved gum from the mucilage were removed when strained with a neat calico cloth. Absolute ethanol was used to precipitate the gum from the solution. The precipitated gum was filtered and washed with diethyl ether and was dried in a hot air oven at 50°C for 48 hours to achieve a dry polymer that could be stored. The TM gum was pulverized again with a blender and stored in an airtight container.

2.4 Preparation of Xanthan Gum, gum Arabic and Terminalia Mantaly polymer

The gum Arabic powder and Terminalia Mantaly powder in various quantities were dissolved in deionized water to get various concentrations, ranging from 5,000 ppm to 10,000 ppm and 20,000 ppm. In percentage, these were 0.5%, 1.0%, and 2.0%, respectively. For the Xanthan gum powder, the same was done but for 2,500 ppm, 5,000 ppm and 10,000 ppm, which represented 0.25%, 0.5% and 1.0%, respectively. A plastic bottle was used in all the solution mixing. Deionized water was measured at 350 ml in each bottle. To determine the exact amount of polymer powder, a spatula was used to scoop the polymer powder into the filter paper, which was then placed on an electronic weighing balance. For accuracy's sake, the fans were turned off in the laboratory to determine the exact quantity. The solutions were stirred gently to achieve homogeneity and then vigorously using a Hamilton Beach Mixer (41). The solution is allowed to stand for 24 hours to allow for complete hydration. Rheological characterization was carried out using an OFITE® Viscometer to determine the rheology with 8 precisely regulated test speeds (shear rates in RPM) of 3 (Gel), 6, 30, 60, 100, 200, 300, and 600. The tests lasted for about 48 hours for each sample. After the first 24 hours, the rheological characterization of the polymer solutions was determined, and after the second 24 hours, the effects of monovalent ions (Na⁺) from NaCl and divalent ions (Ca²⁺) from CaCl₂ were determined and comparisons were made to determine their effects.

3. RESULTS AND DISCUSSION

3.1 Xanthan Gum Concentration and Presence of Ions

Figure 3 shows the effect of increasing the concentration of Xanthan Gum in the absence of salt. No monovalent or divalent ions are present. This explains the rheology of Xanthan Gum in the absence of salinity. As shown in Figure 3, the higher the polymer concentration from 0.25% to 1.0%, the higher the viscosity of the solution at all shear rates. Higher XG concentration also increases the degree of shear thinning. This agrees with earlier works by (24, 41-42, 54). From the values of viscosity, it is evident that XG is excellent for mobility control in the reservoir. In polymer flooding, high viscosity like that of XG is needed for good mobility control. Hence, XG is a good biopolymer except for the associated cost (43).

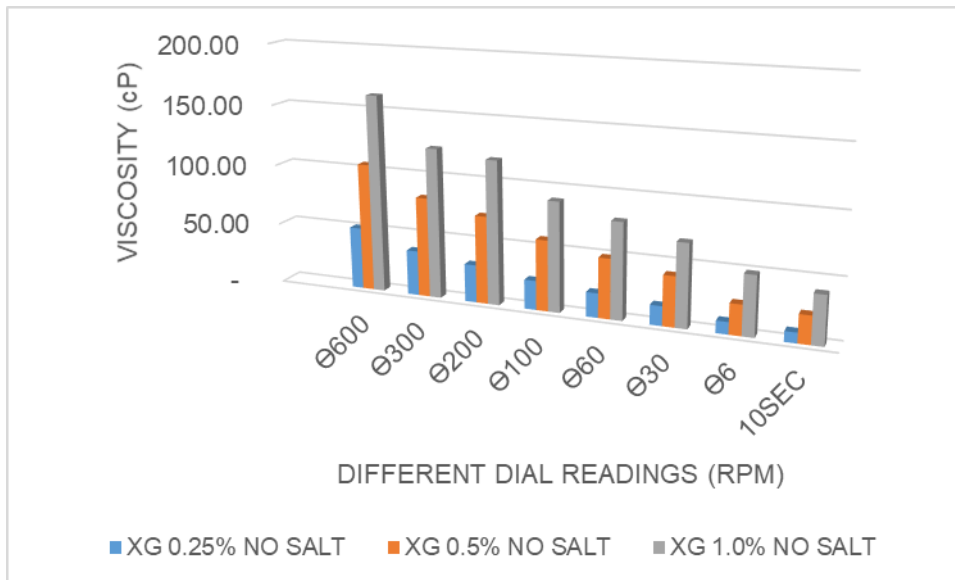


Fig. 3. Effect of XG Concentration with no Salt for 0.25%, 0.5% and 1.0%.

Figures 4, 5, and 6 show the effects of different Xanthan Gum solution concentrations in the presence of 0.75% NaCl, depicting a low salinity reservoir, and 3.5% NaCl and 2.0% CaCl₂, depicting high salinity reservoirs of monovalent ions and divalent ions, respectively. Generally, XG shows good resistance to high salinity irrespective of the concentration of the salt or whether the contribution is monovalent or divalent ion. This is possible because Xanthan Gum's molecular structure has a single, double, or triple helix conformation (24).

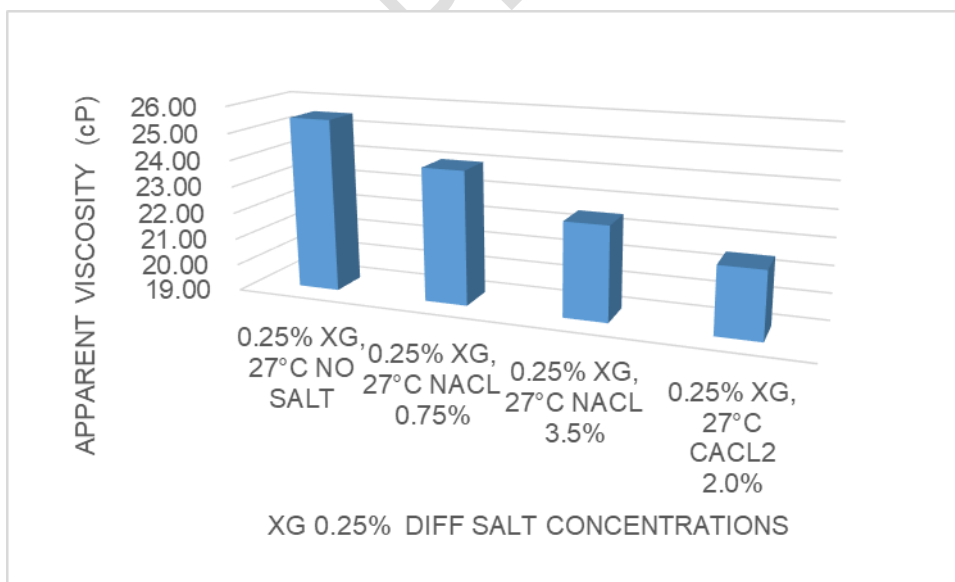


Fig. 4. Effect of Monovalent and Divalent Ions on 0.25% XG Concentration

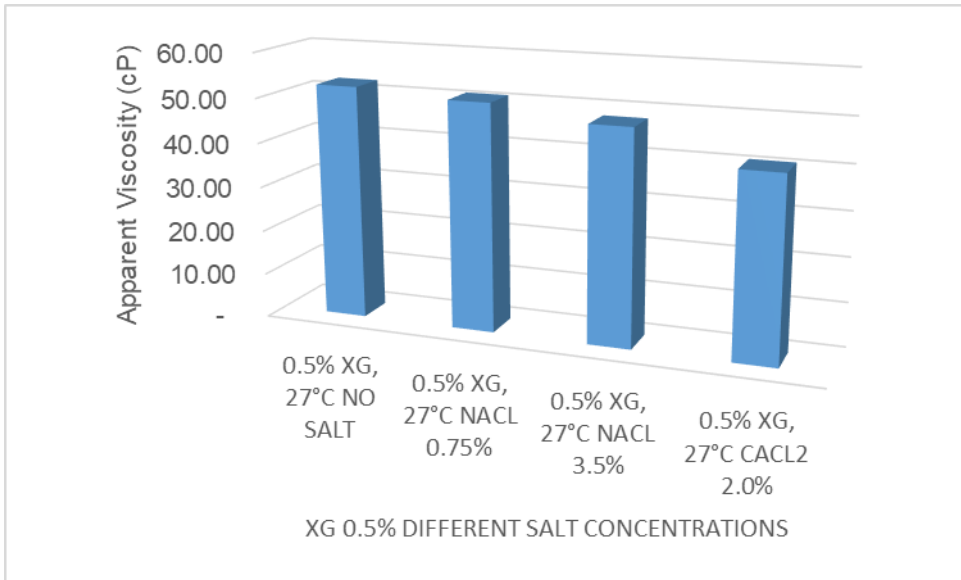


Fig. 5. Effect of Monovalent and Divalent Ions on 0.5% XG Concentration

Figure 6 shows the effect of monovalent and divalent ions on a 1.0% XG concentration. The apparent viscosity of XG 1.0% with no salt and XG 1.0% with NaCl 0.75% are the same. At this concentration, the XG solution is insensitive to the salt that is present. The presence of salt was only felt for XG 1.0% with CaCl₂ 2.0%.

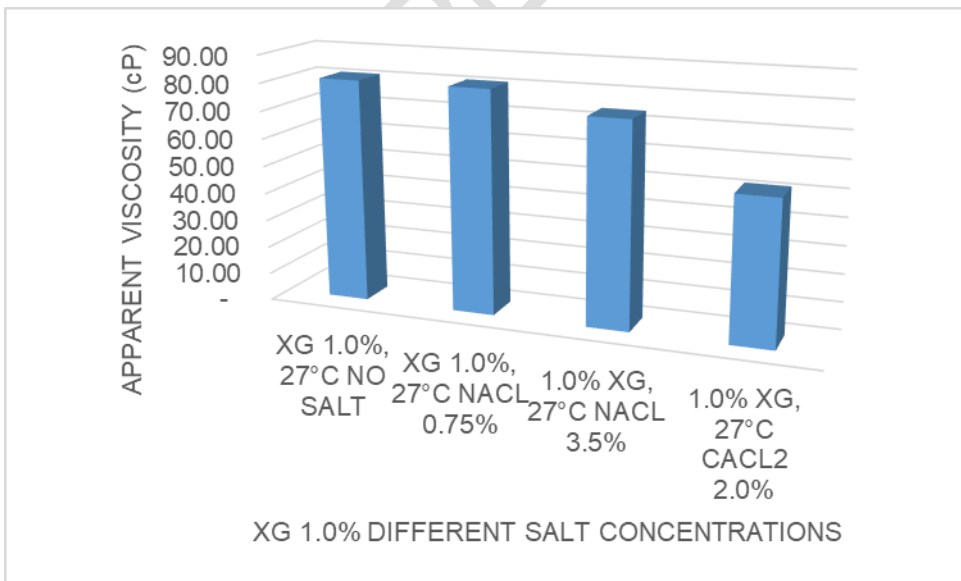


Fig. 6. Effect of Monovalent and Divalent Ions on 1.0% XG Concentration

3.2 Gum Arabic Concentration and Presence of Ions

Figure 7 shows the effect of increasing the concentration of gum Arabic in the absence of salt. No monovalent or divalent ions are present. This explains the rheology of gum Arabic in the absence of salinity. As shown in Figure 7, the higher the polymer concentration from 0.5% to 2.0%, the higher the viscosity of the solution, especially at 600 RPM. At 300 RPM and 100 RPM, the viscosities of both 0.5% and 1.0% are almost the same. At 60 RPM, the viscosities of all GA concentrations are the same. This could be because of the complex arabinogalactan (AG) of GA containing 2% protein (44). From the values of viscosity, it is evident that GA does not form a high viscosity, even at high concentration. This can be an advantage if polymer flooding is designed properly, because there will not be plugging issues. GA is readily available. It is low-cost and biodegradable (45).

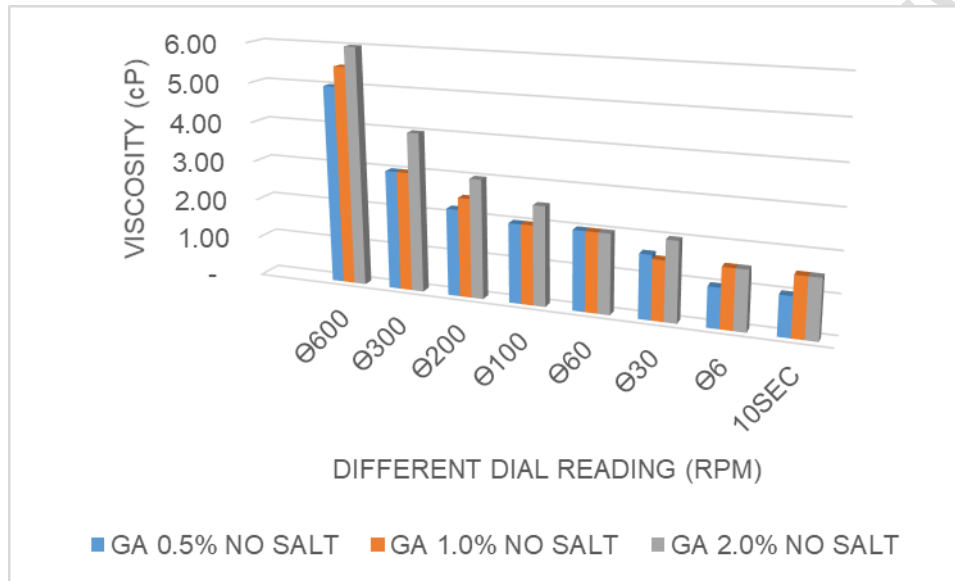


Fig. 7. Effect of GA Concentration with no Salt for 0.5%, 1.0% and 2.0%.

Figures 8, 9 and 10 show the effects of different gum Arabic solution concentrations in the presence of 0.75% NaCl, depicting a low salinity reservoir, and 3.5% NaCl and 2.0% CaCl₂, depicting high salinity reservoirs of monovalent ions and divalent ions, respectively. GA displayed some strange rheological properties. Ordinarily, it is expected that as the salt concentration gets higher, the viscosity of GA in solution should reduce, but this is not so for monovalent ions at 3.5% for both 0.5% and 1.0% GA, where a slight increase in viscosity was noticed. This accounts for the good properties of GA in high monovalent ions. However, as expected at 2% CaCl₂, there was a reduction in viscosity.

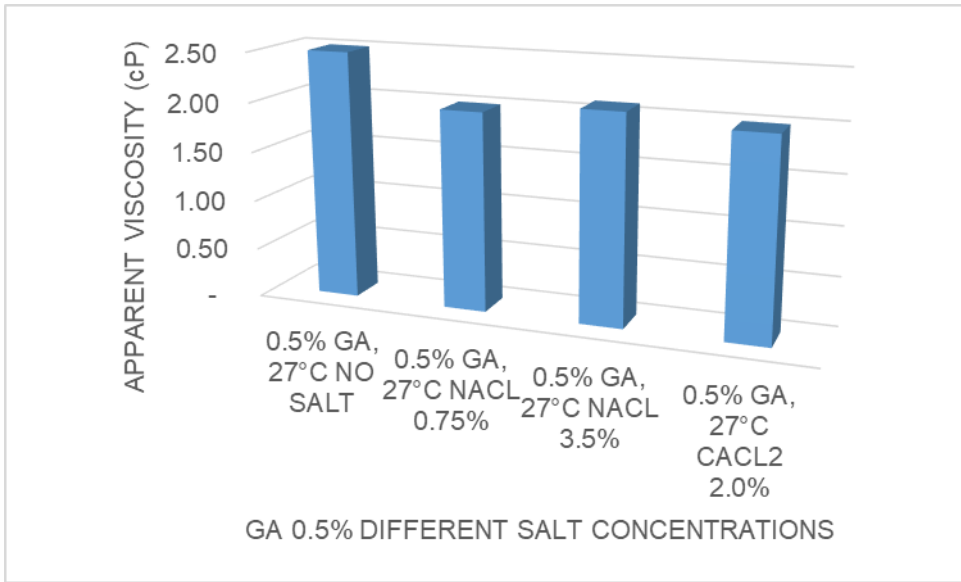


Fig. 8. Effect of Monovalent and Divalent Ions on 0.5% GA Concentration

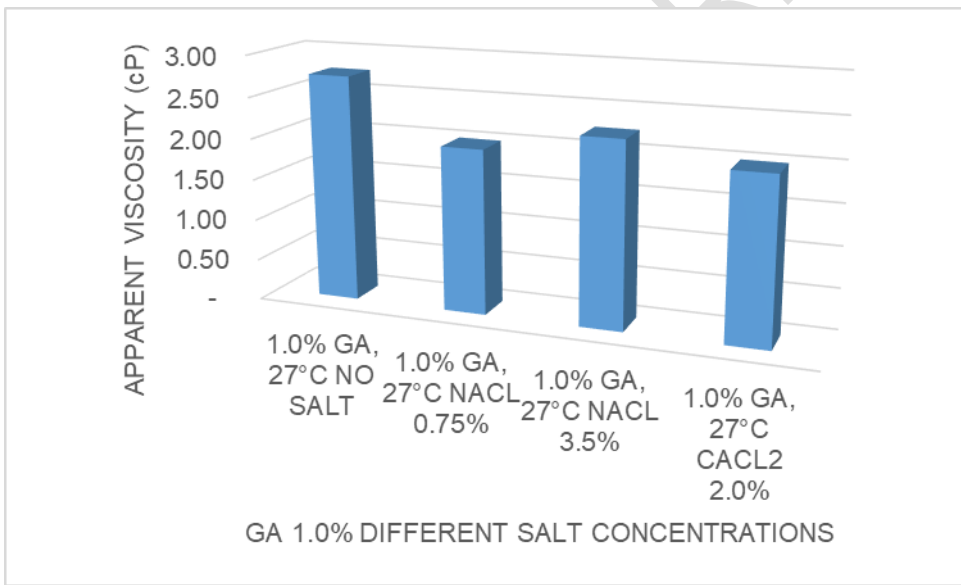


Fig. 9. Effect of Monovalent and Divalent Ions on 1.0% GA Concentration

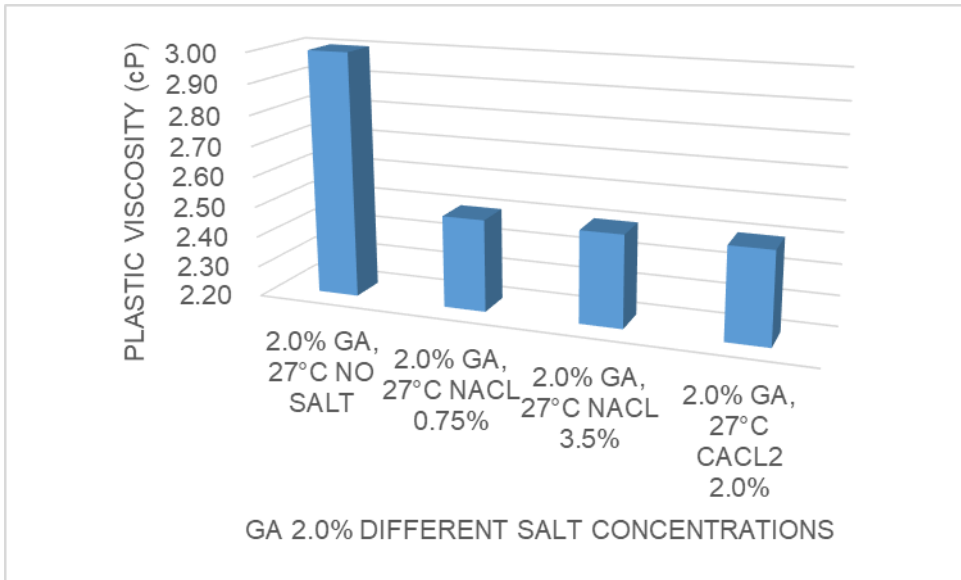


Fig. 10. Effect of Monovalent and Divalent Ions on 2.0% GA Concentration

3.3 Terminalia Mantaly Concentration and Presence of Ions

Figure 11 shows the effect of increasing the concentration of Terminalia Mantaly in the absence of salt. No monovalent or divalent ions are present. This explains the rheology of Terminalia Mantaly in the absence of salinity. As shown in Figure 11, the higher the polymer concentration from 0.5% to 2.0%, the higher the viscosity of the solution. At RPM of 300 and RPM of 200, the viscosities of 0.5% and 1.0% are the same. At RPM's of 100, 30, 6, and 10 sec, the viscosities of 1.0% and 2.0% are the same.

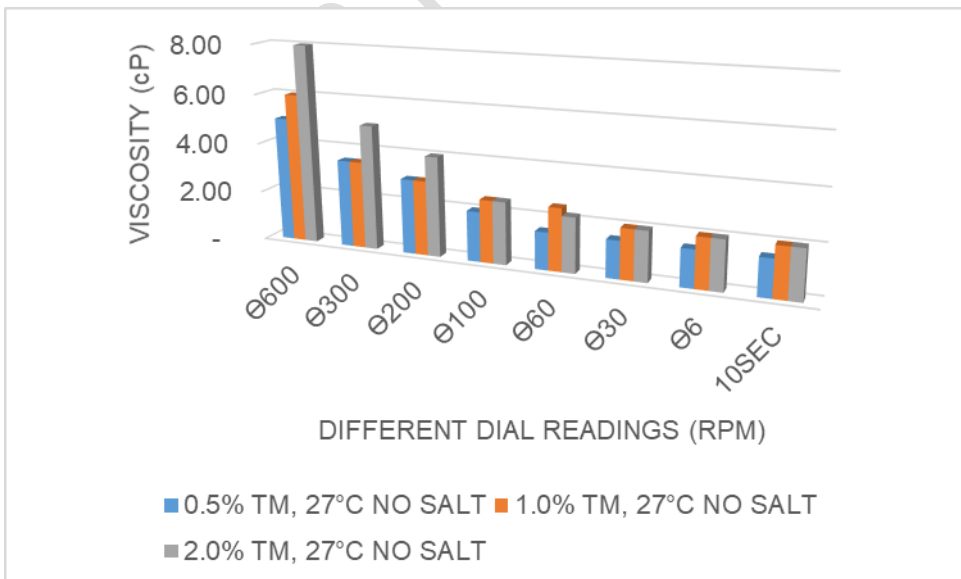


Fig. 11. Effect of TM Concentration with no Salt for 0.5%, 1.0% and 2.0%.

Figures 12, 13 and 14 show the effects of different Terminalia Mantaly solution concentrations in the presence of 0.75% NaCl, depicting a low salinity reservoir, 3.5% NaCl and 2.0% CaCl₂ depicting high salinity reservoirs of monovalent ions and divalent ions, respectively. TM displayed a unique rheological property. Ordinarily, it is expected that as the salt concentration gets higher, the viscosity of TM in solution should reduce, but this is not the case. For all the concentrations, the apparent viscosities were insensitive to salinity. This happened in both the monovalent and divalent ions. Figures 12, 13, and 14 have equal heights, showing the same apparent viscosity for all. This is consistent with earlier works by (39). This is because of the stable repulsive charge polyelectrolyte of the polymer when salinity increases.

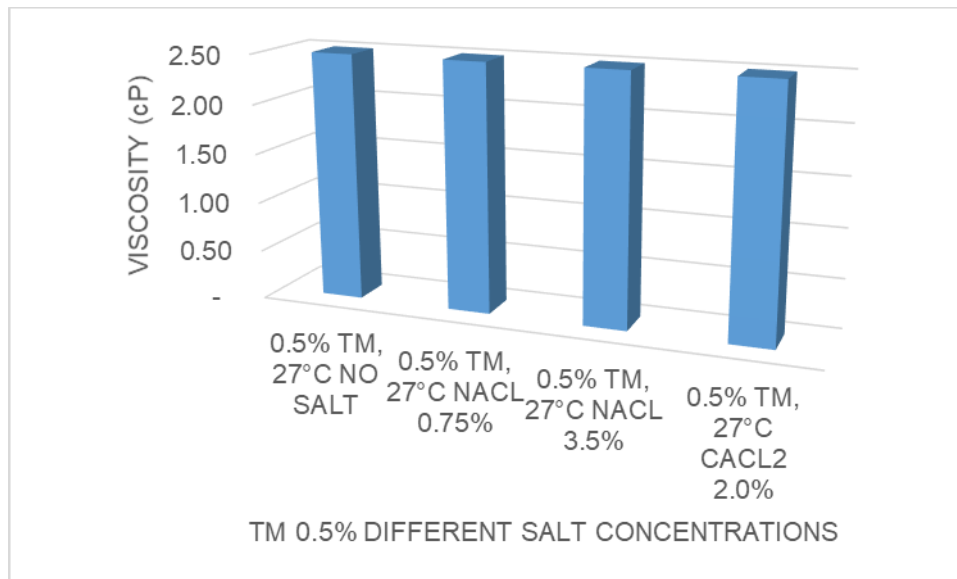


Fig. 12. Effect of Monovalent and Divalent Ions on 0.5% TM Concentration

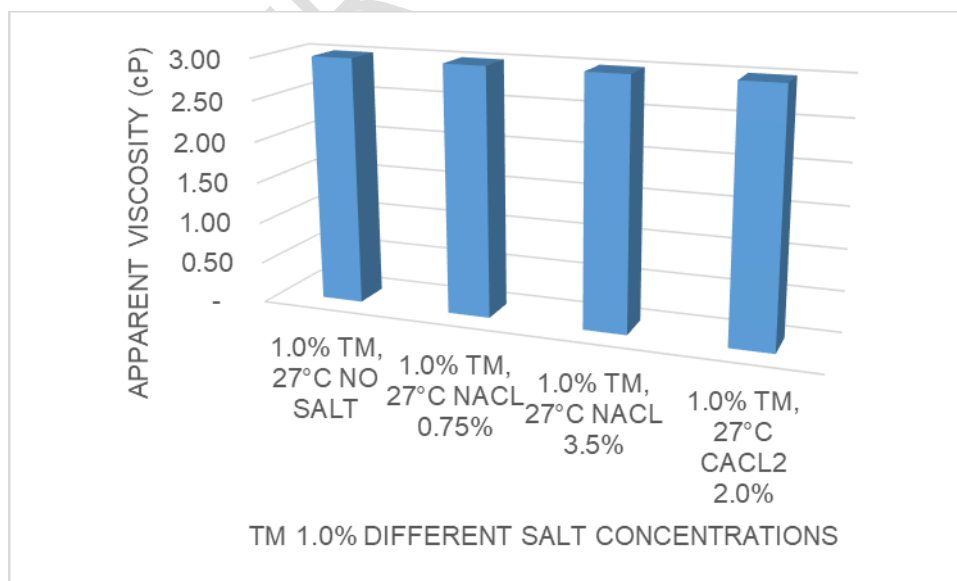


Fig. 13. Effect of Monovalent and Divalent Ions on 1.0% TM Concentration

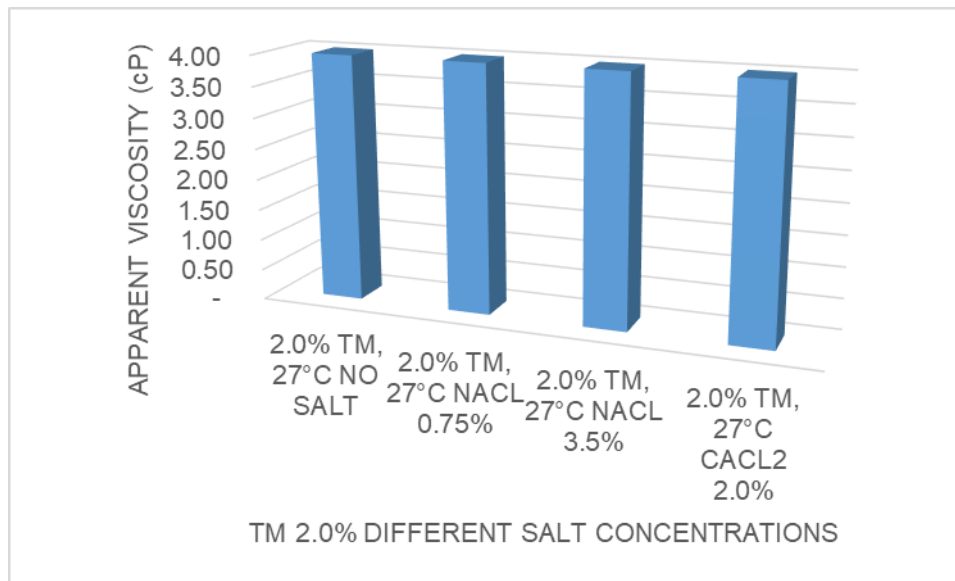


Fig. 14. Effect of Monovalent and Divalent Ions on 2.0% TM Concentration

4.0 CONCLUSIONS AND RECOMMENDATION

The following conclusions and recommendations were reached during the project work:

4.1 Conclusions

- * Even at low concentrations, xanthan gum has a high viscosity in solution.
- * Xanthan gum demonstrated increased stability, particularly at low monovalent ion concentrations (0.75% NaCl). For a high concentration of monovalent ions (3.5% NaCl), XG exhibited increased stability at a high concentration of XG (1%).
- * Gum Arabic also showed increased stability as concentration was increased, but GA had the lowest viscosity of the three biopolymers (XG, GA, and TM). Ordinarily, it is expected that as the salt concentration gets higher, the viscosity of GA in solution should reduce, but this is not so for monovalent ions at 3.5% for both 0.5% and 1.0% GA, where a slight increase in viscosity was noticed.
- * Terminalia Mantaly gum is very resistant to salt, both monovalent and divalent ions. For all the concentrations of TM and salinity, no reduction in apparent viscosity was noticed.
- * The effect of divalent ions is felt more in reservoirs than the effect of monovalent ions.

4.2 Recommendations

* It is recommended that Terminalia Mantaly be investigated more, as it can replace imported biopolymers for Enhanced Oil Recovery (EOR).

* Gum Arabic can be improved upon to withstand salinity like Terminalia Mantaly.

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ABBREVIATIONS

AG	= <i>Arabinogalactan</i>
CaCl ₂	= <i>Calcium Chloride</i>
Ca ₂₊	= <i>Calcium Ions</i>
cEOR	= <i>Chemical Enhanced Oil Recovery</i>
CHCl ₃	= <i>Chloroform</i>
cP	= <i>Centipoise</i>
EOR	= <i>Enhanced Oil Recovery</i>
GA	= <i>Gum Arabic</i>
GP	= <i>Glycoprotein</i>
g/cm ³	= <i>Gram per cubic Centimetre</i>
g/mol	= <i>Gram per Mole</i>

<i>HAPAM</i>	= <i>Hydrophobically Associated Polyacrylamides</i>
<i>HEC</i>	= <i>Hydroxyethyl cellulose</i>
<i>HPAM</i>	= <i>Hydrolyzed Polyacrylamide</i>
<i>HTHS</i>	= <i>High Temperature High Salinity</i>
<i>HSLT</i>	= <i>High Salinity Low Temperature</i>
<i>Kg/mol</i>	= <i>Kilogram per Mole</i>
<i>KYPAM</i>	= <i>Comb Shaped Modified HPAM-A high Salinity Tolerant Polymer</i>
<i>NaCl</i>	= <i>Sodium Chloride</i>
<i>Na⁺</i>	= <i>Sodium Ions</i>
<i>pH</i>	= <i>potential of hydrogen</i>
<i>PPM</i>	= <i>Parts Per Million</i>
<i>RPM</i>	= <i>Revolution per Minute</i>
<i>TM</i>	= <i>Terminalia Mantaly</i>
<i>XG</i>	= <i>Xanthan Gum</i>
<i>µm</i>	= <i>Micro Metre</i>

UNDER PEER REVIEW