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Hydrophobic Associating Amphiphilic Tetrapolymer for Improved Stability of Drilling Mud

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Abstract

The exploration for functional polymers to improve the performance of water-based drilling muds is work in progress given the variations in temperature, pressure and salinity of formations during wellbore development. This work synthesised hydrophobic associating amphiphilic tetrapolymer (HAAT) via micellar polymerization. The chemical structure and properties of the polymer were characterized by FTIR and ^{1}H NMR spectroscopy, intrinsic viscosity test, thermogravimetric analysis (TGA) and differential thermal analysis(DTA). TGA and DTA established that the HAAT polymer had thermal resilience up to about 343.0°C. The performance of the HAAT polymer compared with KPAM as additives for enhancing the rheological quality and filtration loss control of aqueous bentonite-based unsalted mud and barite-weighted bentonite-based salted-mud were assessed. Both improved the temperature stability and salt tolerance as reflected in the preservation of rheological properties measured after high-temperature treatment of the drilling mud with HAAT polymer showing superiority by better-retention of mud parameters. This was further confirmed by excellent fluid retention of the muds under both standard API and high-temperature high-pressure (HTHP) filtration conditions as both HAAT and KPAM demonstrated at least six-fold fluid loss control at 150°C and 3500 KPa compared to the blank.

Keywords: *HAAT tetrapolymer; Characterization; Drilling mud; High temperature; Mud properties.*

1. Introduction

Drilling fluids are exceptionally essential in rotary boring activities of oilwells. They are generally circulated down through the hollow drill string, exit at the bit nozzles and return through the annulus to the surface. The complexity of mud composition depends on the type of formation, formation pressure, the hole depth and temperature among others. Water may be adequate during the initial stage of drilling, particularly close to the surface, but more complicated mud is required at greater depths. Drilling fluids (muds) can be classified into water-based muds (WBDF), oil-based muds (OBDF), synthetic-based drilling fluids (SBDF) and gas-based drilling fluid (GBDF) [1-2]. Among these fluids, water-based muds are generally preferred because they are environmentally friendly and relatively cheaper, basically credited to the copious use of water as the dispersing phase [3]. The muds serve multifunctional purposes for successful drilling operations and should exhibit minimal interactions with the formations for the integrity of the open hole [4]. Prominent among the functions are provision of hydrostatic pressure, cooling and lubricating of the drill bit and transporting of rock cuttings and cleaning of hole, fluid loss control and mechanical stability [5-6]. The ability of the mud to perform these functions requires some minimum stability of the most vital mud components and properties [7].

Mud density provides hydrostatic pressure against the formation pressures and buoyancy for particle suspension. Muds derive their weights mainly from the base fluids and weighting materials which are either soluble salts or insoluble solids [8]. The mud weight can be adjusted upward using weighting materials or adjusted downward using water. In clay-free aqueous mud, filtration loss cannot be controlled without the presence of water-soluble polymers which is a necessary additive for viscosifying the base fluid mainly for fluid loss control rather than suspension of weighting agents. For clay-suspended water-based muds, both insoluble solids and soluble salts add weight to the muds. The addition of polymers is crucial to provide viscosity, maintain the insoluble solids suspension, promote fluid loss control, and promote cuttings transportation and suspension.

Filtration property is a very important characteristic of muds as it is an indication of the loss of the base fluid into the porous formations [9]. Loss of fluids to the porous formations cause formation damage resulting from rock wettability changes, fines migration, clay swelling, fluid solids plugging and formation water chemistry incompatibilities [10]. Filtration loss is usually minimized by the addition of fluid loss control agents. Most common agents that provide viscosity and fluid loss control are clays, especially bentonite clay, and water-soluble polymers.

Bentonite clay is a common ingredient used for mud primary viscosity enhancement and fluid loss control because it swells considerably and favourably in the presence of water. Additionally, its clay particles can plug pores and fractures in the vicinity of wellbore, thus, forming thin and low-permeability filtration cakes capable of reducing filtration losses. However, at high-temperature, high chemical contamination or both, low bentonite water-based muds experience excessive gelling, suffer dehydration and flocculation resulting in drastic filtration loss [5,11-12]. High content bentonite water-based drilling fluids equally experience deterioration of fluid properties under such unfavourably wellbore conditions [13-14]. To strengthen the viscosity and filtration loss control in aqueous muds, the addition of water-soluble polymers is recommended [2,15]. Nonetheless, some drawbacks of most commonly used polymers for petroleum resource recovery are degradation at elevated temperatures and deterioration under high saline media [16]. Natural polymers which are more ecofriendly and salt tolerant but are prone to oxidation and bacterial action [17-18]. Synthetic homopolymers derived from acrylamide monomers are susceptible to depreciation when subjected to high-salinity, low pH, high shear rate, and high-temperature [19].

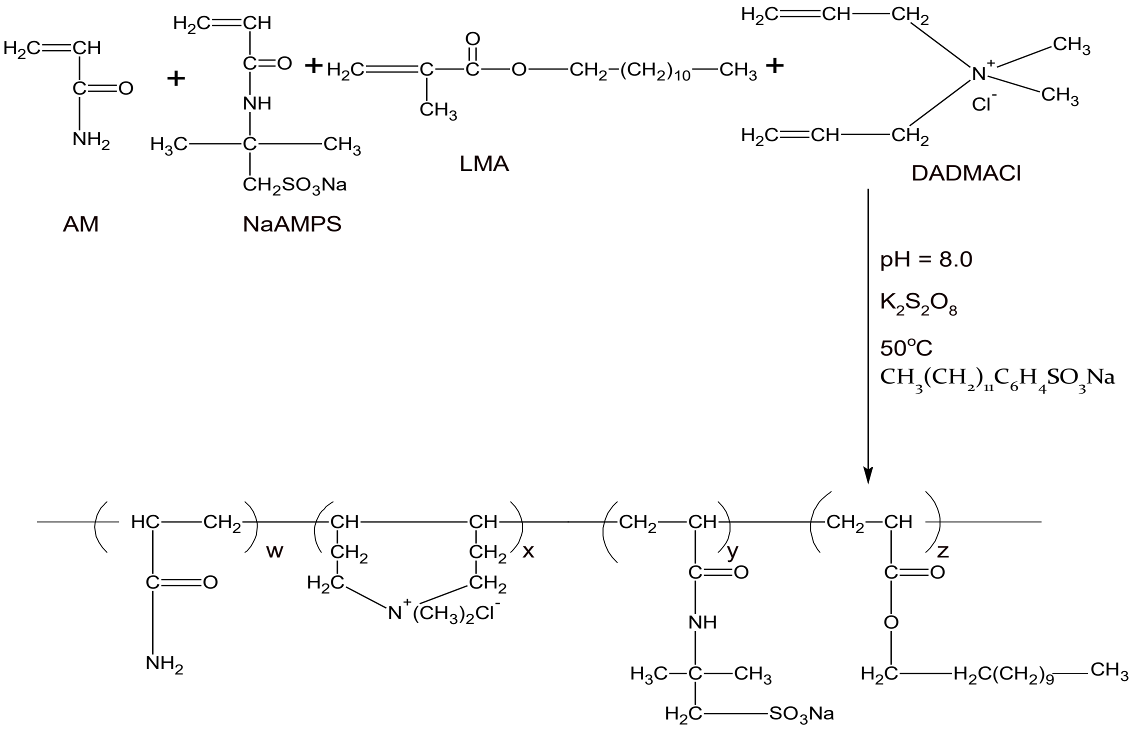
It is imperative to develop copolymers to improve the performance of low content bentonite water-based mud under the influence high temperature resistance, high salinity or both. A HAAT polymer was designed to exhibit thermal and chemical stability for applications in mud. The amide group of AM monomer which forming the main chain structure enhances viscosity. The sulfonate side group of AMPS promotes hydration and strengthens the rigidity of the polymer chain to improve thermal stability of the product. The hydrophobic lauryl methacrylate monomer will impart hydrophobic associations in saline environment to reduce the salt screen that tends to reduce the viscosity ionic polymers in aqueous media. The cationic divinyl monomer (diallyldimethylammonium chloride) introduces cyclic and crosslinked or mesh structures during polymerization. The cyclic structures potentially provide rigidity and steric hindrance against thermal degradation and viscosity loss in saline medium.

2. Experimental, materials and synthesis of HAAT tetrapolymers

Materials used were acrylamide (AM, 98.0%), 2-acrylamido-2-methylpropanesulfonic acid (AMPS, ≥ 90%), lauryl methacrylate (LMA, 96.0%), diallyldimethylammonium chloride (DADMACl, 60.0% in water), sodium dodecylbenzene sulphonate (90.0%), 2-2'-azobisisobutyronitrile (AIBN, 94.5%), potassium persulphate ($K_2S_2O_8$, 99.50%), sodium metabisulphite ($Na_2S_2O_5$, 96.0%), sodium hydroxide (96.0%), sodium chloride (99.5%), deionized water, acetone (99.5%), ethanol (99.7%).

The HAAT polymer was synthesised by micellar polymerization using mole ratios of 69%AM/27% AMPS/1.3%LMA/2.7% DADMACl, based on previous report[20]. Reaction parameters were total monomers concentration of 21.8 wt/vol.%, pH=8.0, at 50°C and AIBN

0.5wt%, 2.0% sodium dodecylbenzene sulphonate (surfactant) and reaction time of 1.0 hr. Firstly, AMPS was dissolved in deionised water and 2M NaOH added to adjust the pH. Acrylamide dissolved in deionised water was added to AMPS solution, thoroughly mixed and the pH adjusted before transferring it into the three-neck flask and purging with nitrogen for at least 20 minutes. While stirring the mixture, lauryl methacrylate, diallyldimethylammonium chloride and sodium dodecylbenzene sulphonate (SDS) were mixed proportionally and added, followed by the initiator. After reaction, the HAAT tetrapolymer as presented in Scheme 1 was isolated from the highly viscous mass using 50:50v/v acetone-ethanol solvent system in excess and the product dried at 60°C.



Scheme 1.

3. HAAT structures and functional properties

The HAAT was designed to improve viscosity and stability of muds under influence of relative high temperature or saline environment or both. Structurally, the sulphonate side chain of AMPS, dimethyl and carboxylate groups can contribute thermal resistance. The hydrophobic lauryl methacrylate monomer will impart hydrophobic associations in saline environment to retain the polymer viscosity. The cationic divinyl monomer (diallyldimethylammonium chloride) enrich polyelectrolytic function, introduces crosslinking structures in the polymer. The pendent-chains incorporate comblike-structures which provide rigidity and steric hindrance against thermal degradation and viscosity loss in saline medium. The intrinsic viscosities $[\eta]$ and viscosity-average molecular weight of HAAT copolymer (M_V) were determined as 9.36 dL/g and 4.64 x10^6 g/mol, respectively.

4. HAAT and mud properties evaluation

Fourier transform infrared (FTIR) radiation and proton nuclear magnetic resonance (^{1}H NMR) spectra and thermogravimetric and differential thermal analyses (TGA, DTA) of the HAAT polymer were determined and interpreted according to previous reports [4,21].

Muds were formulated and the rheological properties and fluid losses determined according to American Petroleum Institute (API) procedure [22] and previous reports [4,20-21].

5. Results and discussion

5.1. Characterization of HAAT polymer

5.1.1. FTIR and ^{1}H NMR spectra of HAAT polymer

The HAAT structure was confirmed based on the following FTIR spectral analysis. As shown in Fig. 1a, there were absorptions at 3434.67cm^{-1} from –NH stretching and 2934.52 cm^{-1} from –CH_2 stretching vibration, 1661.56 cm^{-1} from C=O stretching vibration, and 1543.56 cm^{-1} (C–N stretching vibration and –NH bending vibration. The -N-C symmetrical stretch of DADMACl occurred at 1116.67 cm^{-1}-1189.14 cm^{-1} and overlapped with asymmetric stretching of the sulfonate group at 1189.14 cm^{-1}. A very sharp signal recorded at 1042.00 cm^{-1} reflects symmetrically stretched sulphonate from AMPS. C-H bonds of -CH_3 group rocking occur at 1390.86 cm^{-1}. Furthermore, peaks at 1455.06 cm^{-1} were probably from methyl or methylene bending while 813.00 cm^{-1} and 688.73 cm^{-1} confirmed the alkane backbone of the tetrapolymer. As expected, the FTIR spectrum has established the polymerization of the four monomers to form HAAT. Fig. 1b shows the ^{1}H NMR spectrum of HAAT, proton signals that resonate at chemical shift, 1.2, 1.5 ppm indicated methylene (-CH_2) on the tetrapolymer backbone chain, and -CH_2 protons on the ring of DADMACl and along the side chain of LMA, in addition to methyl (–CH_3) on the AMPS and LMA. The methine proton (-CH) of the backbone chain of HAAT appeared at δ =2.1. The hydrogens of the -CH_2 group bonded to sulphonate were noticed at 3.4 ppm. The chemical shift at δ = 3.6 ppm reflected hydrogens in -CH_2-N(CH_3)$_2$Cl-CH_2- of DADMACl and the methoxy (-CH_2O-) of LMA. The peak at 4.7 ppm was accredited to the (H) from -NH and -NH_2 and D_2O.

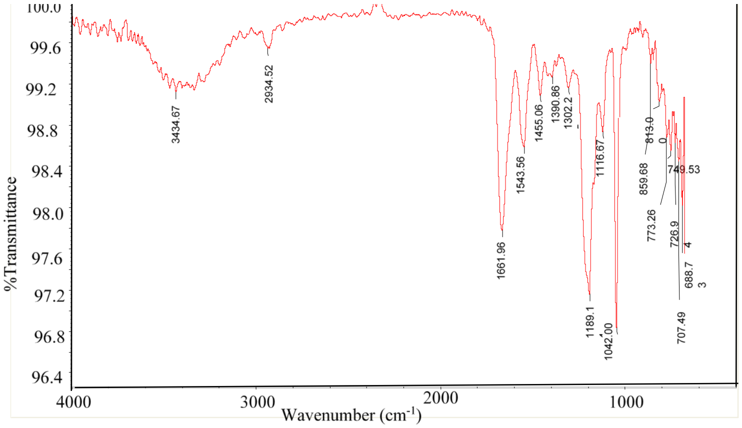


Fig. 1a FTIR spectrum of HAAT polymer.

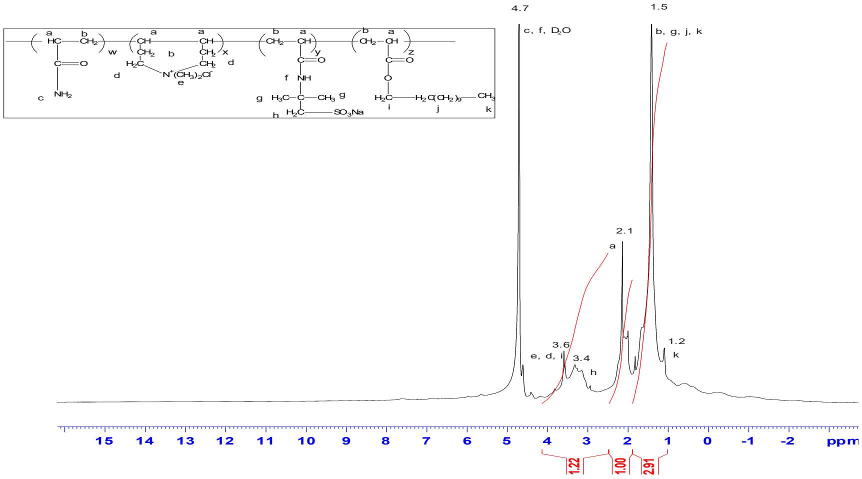


Fig. 1b ^{1}H-NMR spectrum of HAAT polymer.

5.1.2. Thermal analyses of HAAT polymer

Thermogravimetric and differential thermal analyses as presented reflect the thermal properties and behaviour of HAAT polymer when subjected to heat treatment. On the TGA curve in Fig. 2, the initial gradual decline indicates moisture evaporation and volatile matter loss. The sharp substantial change at 343.1°C on both TGA and DTA curves (Fig. 2) reflects weight losses as result of thermal decomposition from emission of gases such as CO_2 (acrylamide), NH_3(acrylamide) SO_2 (2-acrylamido-2-methyl propane sulfonate) and polymer chain cleavage products. The temperature, 343.1 °C marks the upper threshold beyond which the HAAT polymer is not useful as an additive in drilling mud applications. However, this temperature reveals high decomposition temperature and therefore indicates that the polymer has high thermal resistant attributable to the presence of the sulfonate group $—SO_3$, cross-linked and possible cyclic ring structures in it.

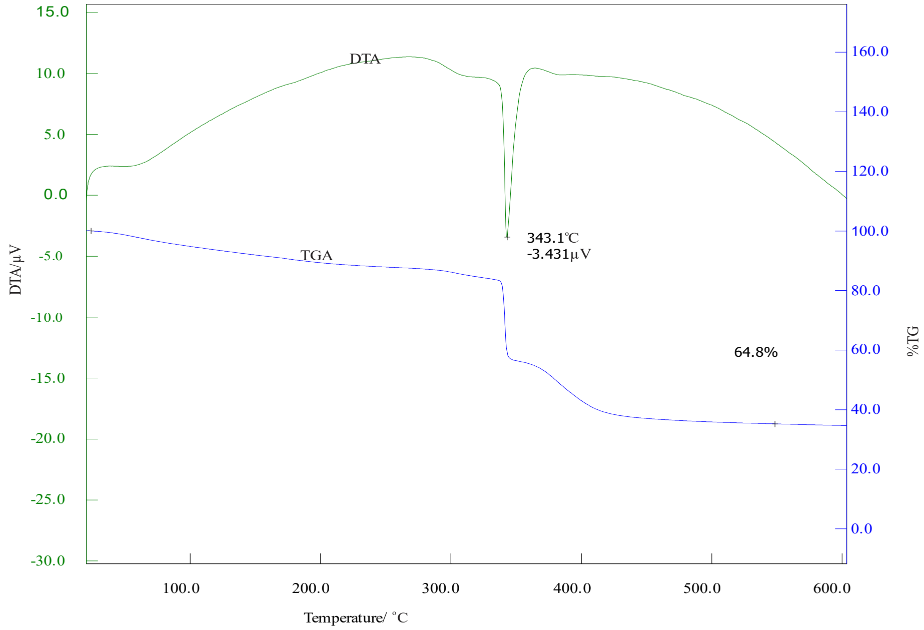


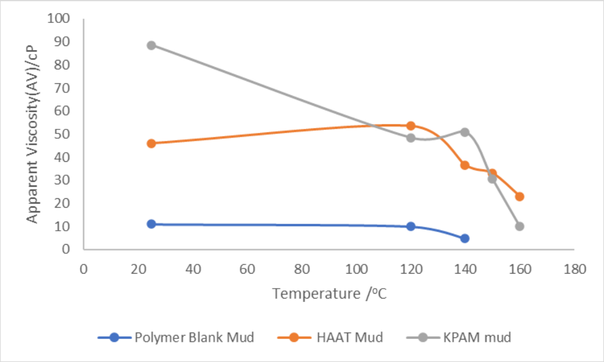
Fig. 2. Thermogravimetric and thermal differential curves of HAAT polymer.

5.2. HAAT polymer effects on mud properties

5.2.1. Apparent viscosity (AV)

Mud viscosity shows the internal resistance to flow and reflects the adhesive and cohesive interactions of dissolved solutes, suspended particles and liquid base media. Shear stress is directly proportional to shear rate for Newtonian fluids where apparent viscosity is constant but varies for non-Newtonian fluids. While low mud viscosity favours the rate of bit penetration, the mud viscosity which determines its hole cleaning and solids suspension efficiency in the annulus should be sufficient viscous to suspend and transport cuttings to the surface but not so high as to minimize frictional pressure losses.

The apparent viscosity of both non-salted and salted-polymer (5% NaCl) muds increased conspicuously when HAAT and KPAM were added to the muds as juxtaposed with the polymer-free-blank (Figs. 3-4). However, considering AV, muds were sensitive to temperature in the range of 120 °C -150°C. For non-salted mud, KPAM-mud experienced a more viscosity loss of 88.7% while HAAT-mud reduced by 50.0% after heating at 160°C with respect to the initial AV values at 25°C (Fig. 3). With salted-muds after heat treatment at 150°C (Fig. 4) when compared to values of unaged salted muds at 25°C, viscosity losses were 8.5% (HAAT), 35.7% (KPAM) and 52.0% for the blank which experienced the highest viscosity loss.



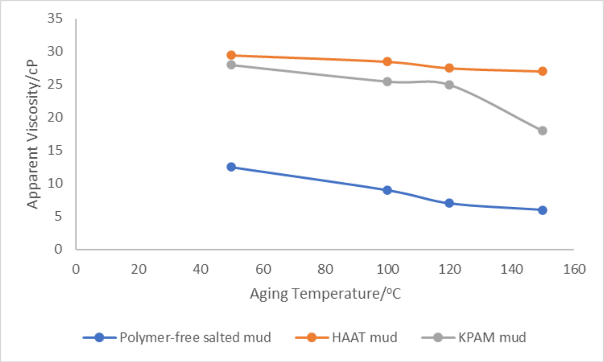


Fig. 3. Effect of temperature on apparent viscosity of polymer-incorporated mud.

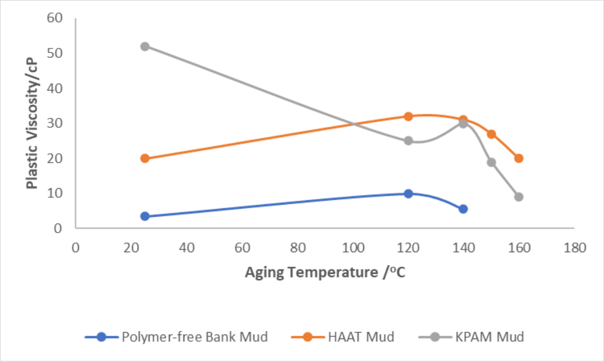
Fig. 4. Effect of polymer addition on apparent viscosity of salted mud at elevated temperatures.

5.2.2. Plastic viscosity (PV)

PV revealed internal friction between fluid layers under shear stress which depends on the concentration, shape and size of solids as well as the base liquid viscosity. During drilling operations, maintaining an optimal plastic viscosity is vital for efficient transport of cuttings and hole cleaning, hydraulic power and pump efficiency, and mud stability and solids suspension. Optimum plastic viscosity improves fluid mobility and lowers energy requirements for pumping. When using a direct indicating viscometer, PV is determined using the relationship below.

Compared to the unaged non-salted mud in Fig. 5, plastic viscosities for non-salted HAAT-mud was retained whilst plastic viscosity KPAM decreased by 82.7% after hot-aging at 160°C. The blank mud PV increased by 57.1% at 140°C, but its yield point disappeared. For salted mud (Fig. 6), plastic viscosity of HAAT-mud and KPAM-mud reduced by 22.2% and 31.3%, respectively, after rolled-heating at 150°C considering PV values of unheated fresh muds. polymer-free blank suffered the worst PV loss by 42.9%.

With reference to only PV, the blank had the least value and can facilitate quicker drillbit penetration the formations, however, its excessive filtration loss and low AV indicating a poor property to suspend solids will render it less effective compared to the polymer- incorporated muds.



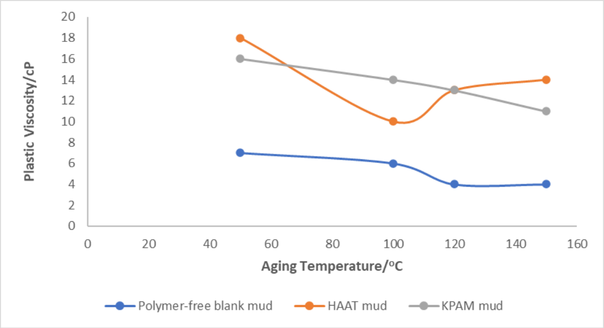


Fig. 5. Effect of Temperature on plastic viscosity of polymer-incorporated mud.

Fig. 6. Effect of polymer additive on plastic viscosity of salt contaminated mud at elevated temperatures.

5.2.3. Yield point and gel strengths

Yield point (YP) is the initial resistance to fluid flow contingent on the electrochemical forces of attraction between the particles. It is responsible for mud gelling under static condition and indicates the minimum stress required for a mud to flow. It reflects surface properties, mud solids concentration and the mud ability to lift cuttings out of the annulus. A high yield point implies a non-Newtonian fluid with better capacity to carry cuttings than a fluid of similar density but lower yield point.

Yield point of KPAM decreased by 97.3% compared to 81.3% for HAAT at 160°C in the non-salted mud. The polymer free mud (blank) however, lost its yield point after hot-aging at 120°C and recorded a negative value at 140°C (Fig. 7). In the case of salted mud, whereas the yield points of the blank decreased by 63.6%, KPAM-mud by 41.7%, HAAT mud rather appreciated by 13.0% (Fig. 8).

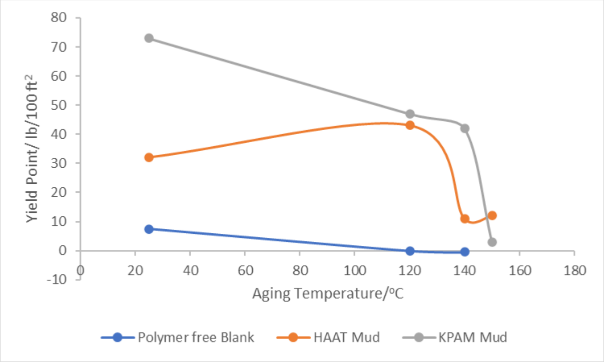


Fig. 7. Effect of Temperature on yield point of low solids bentonite-based mud.

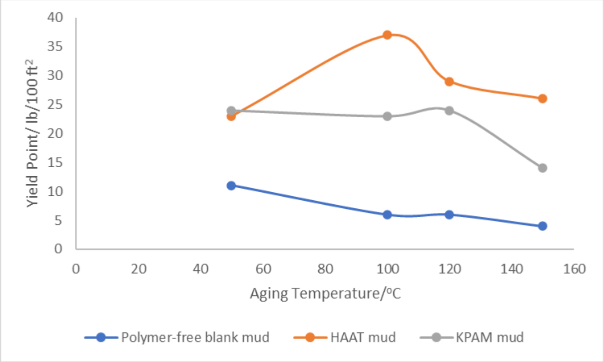


Fig.8. Effect of polymer additive on yield point of salt contaminated mud at elevated temperatures.

Moderately high retention of yield strengths of the muds after heat-aging, particularly, because of the addition of the polymers, demonstrated better effectiveness of the HAAT-aided mud to convey cuttings than the blank mud with comparable weight but lower yield point.

Gel strength of drilling fluids is attributed to the presence of electrically charged molecules and solids such as clay particles which agglomerate into a firm matrix when circulation stops. and usually subject to chemical treatment, solids concentration, time, and temperature. It represents shear stress (expressed in lb$_f$/100 ft^2) and determined by the maximum dial deflection when the rotational viscometer is set to low shear rate, usually at 3 rpm, after a mud has set quiescently for a period of time, generally, for 10-seconds or 10-minutes by API procedure [23-24]. As shown in Fig. 9, KPAM effect in the unsalted mud showed a wider difference between the initial and 10-minute gel strength which indicated progressive gelling which is less desirable compared to the HAAT and the blank. For salted muds in Fig. 10, while the blank provided relatively the lowest gelling, HAAT effected fairly stable gel strength whereas KPAM was more sensitive and responded more adversely to elevated temperature treatment. Generally, gel strength and yield points nearly followed similar patterns since both measure the attractive forces in mud systems and therefore, require similar chemical treatments for modification where necessary. The 10-second gel reading more closely approximates the true yield stress in most drilling fluid systems. Water dilution can be effective in lowering gel strengths, especially when solids are high in the mud.

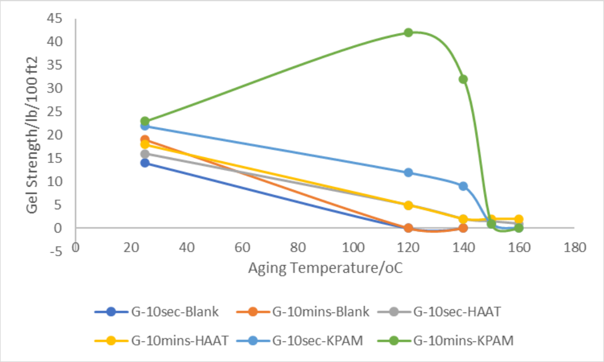


Fig.9. Effect of polymer additive on gel strength of mud at elevated temperatures.

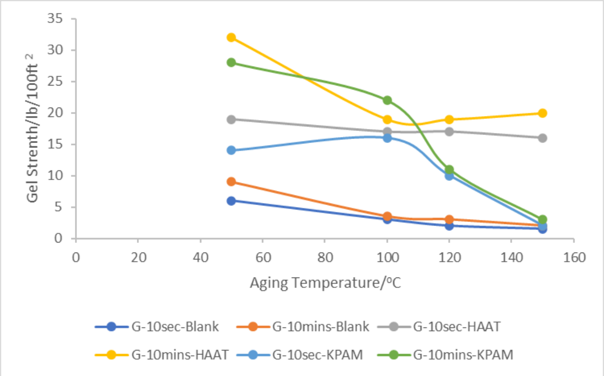


Fig. 10. Polymer effect on gel strength of salted mud exposed to high temperatures.

5.2.4. Filtration loss

Fluid filtration quantities determined by standard API and HTHP procedures indicate the volumes of drilling mud that can seep into the formation during drilling. Filtration loss is affected by mud properties, formation characteristics, pressure differential between mud column and formation and filter cake quality and thickness It is a vital parameter in drilling operations, as excessive fluid loss causes formation damage, pipe stucking and reduction in mud effectiveness.

Filtration losses of the unaged and after hot-aging of the muds at 25 °C, 120°C-160°C are presented in Fig. 11. The filtration loss of KPAM-mud increased by 37.5% at 160°C. Comparatively, HAAT maintained nearly constant fluid loss with an average value of 10.5 cm^3/30 min. Filtration losses in all the mud systems before and after heat treatment between 120°C-160°C were acceptably low However, both KPAM and HAAT exhibited better fluid loss control compared to the blank which suffered excessive increase in fluid loss of about 350% after heating at 140°C.

As presented in Fig. 12-13 regarding fluid losses of salted-muds at varied thermal-aged conditions, for API filtration at 100 psi, blank-mud increased from 43.5cm^3/30min (non-hot aged at 25°C) to 60cm^3/30min after dynamic heat treatment at 150°C whilst the fluid losses (generally below 10.0 cm^3/30 min) of the polymer-muds were weakly affected by heat exposure in the stipulated temperature range. HAAT and KPAM demonstrated significantly high fluid loss reduction by providing at least 6.5-fold and 6-fold, respectively, HTHP fluid loss control at 150°C compared to the blank (113.2 cm^3/30min).

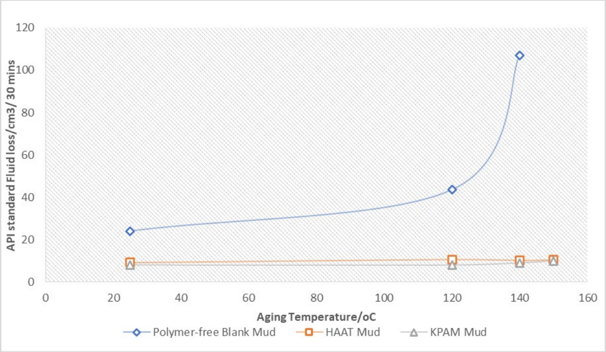


Fig. 11. Polymer effect on filtration of unsalted mud exposed to high temperatures.

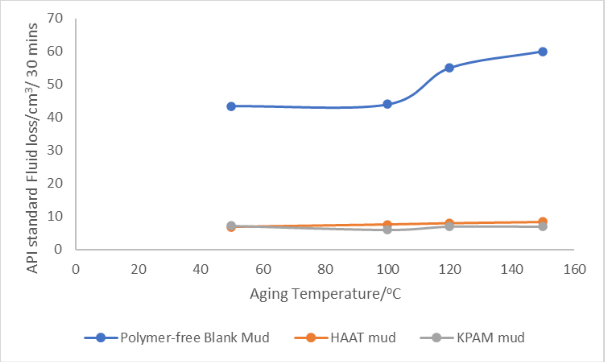


Fig. 12. Polymer effect on filtration of salted mud exposed to high temperatures.

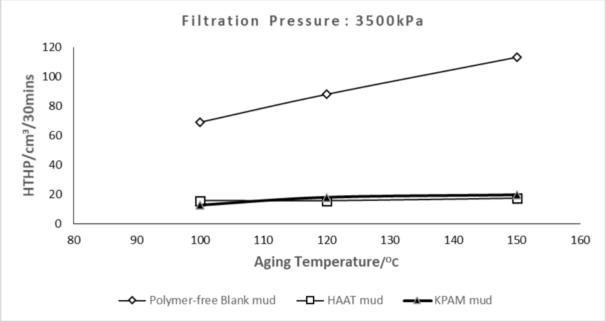


Fig. 13. Polymer effect on filtration of salted mud exposed to high temperature-high pressure.

Presence of polymers in the mud generate two mechanisms that reduce the rate of filtration. Polymers can encapsulate the clay particles through electrostatic interactions thereby creating a thin resilient clay-polymer filter cake which hinders cross-movement of base fluid through the cake. The second mechanism is based on inter/extra polymers molecular interactions and polymer-water associations triggering viscosity enhancement which mitigate water mobility into the clay pores, thereby effecting filtration loss control.

Thus, the more viscous the mud the lower the seepage or fluid loss and the better and thinner the filter cake. With low fluid loss during drilling, the mud properties are less compromised and therefore the mud can perform its key functions for a prolonged period to ensure hole integrity and minimize formation damage.

KPAM mud and HAAT mud showed higher thermal stability and salt tolerance than the blank, with HAAT exhibiting slight superiority. The unrestrained fluid loss of the polymer-free mud indicated, the absence of protection and dispersion-stabilizing agent (polymer), resulting in phase separation and structural breakdown of the bentonite clay and other mud additives. The relatively higher quality of HAAT polymer than KPAM is accredited to structural composition of the latter. The sulphonate side chain of AMPS, dimethyl and carboxylate groups of HAAT contributes its thermal resistance. The hydrophobic lauryl methacrylate monomer imparts hydrophobic associations in saline environment to substantially retain the polymer viscosity. Additionally, there was a moderate increase in association between the hydrophobic groups on different polymer molecules of HAAT with increasing temperature attributed to their hydrophobicity [25]. The cationic divinyl monomer (diallyldimethylammonium chloride) enrich polyelectrolytic function, enriches polyelectrolytic function and introduces in the polymer. The pendent-chains incorporate comblike-structures which provide rigidity and steric hindrance against thermal degradation and viscosity loss in saline medium.

6. Conclusion

A hydrophobic associating amphiphilic tetrapolymer (HAAT) comprising acrylamide, 2-acrylamido-2-methylpropanesulfonate, lauryl methacrylate and diallyldimethylammonium chloride was successfully synthesized via micellar polymerization and characterized. HAAT polymer had thermal resilient threshold of about 343.0°C based on thermogravimetric and differential thermal analyses.

The HAAT polymer had comparable capacity as KPAM to increase the viscosity of aqueous bentonite-based unsalted mud and barite-weighted bentonite-based salted-mud. The HAAT polymer demonstrated relatively higher thermal stability and salt tolerance in the mud than KPAM at 160°C. The HAAT polymer also exhibited excellent fluid retention in the muds under both standard API and high-temperature high-pressure (HTHP) filtration conditions by at least six-fold fluid loss control at 150°C and 3500 KPa compared to the blank. HAAT polymer, thus, has the potential to be an effective substitute for KPAM as a mud additive for viscosity enhancement and fluid loss control.

Abbreviations

AV: Apparent Viscosity, $AV = \emptyset_{600}/2,$ *cP*
PV: Plastic Viscosity, $PV = \emptyset_{600} - \emptyset_{300},$ *cP*
YP: Yield Point, $YP = \emptyset_{300} - PV,$ *lb/100ft*^{2}
HAAT: Hydrophobic associating amphiphilic tetrapolymer
KPAM: Potassium polyacrylamide

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