

THERMOSALINE RESISTANT ACRYLAMIDE-BASED POLYELECTROLYTE AS FILTRATION CONTROL ADDITIVE IN AQUEOUS-BASED MUD

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Abstract

A water-soluble polyelectrolyte comprising acrylamide (AM), sodium salt of acrylic acid (SAA), sodium salt of 2-acrylamido-2-methylpropane sulfonate (SAMPS), and N, N'-dimethyl acrylamide (DMAM) was synthesized by free radical polymerization. Monomeric composition of the polyelectrolyte was established by Fourier transform infrared spectroscopy. The rheology of the polymer in aqueous drilling fluid was evaluated using viscosity, yield point, gel strength, and filtrate loss as performance indicators. Thermal stability and salt tolerance of the polymer-incorporated mud were examined. Furthermore, its salt tolerance in aqueous brine medium was investigated. The polyelectrolyte demonstrated significant improvement in rheological properties and filtrate loss control over a wide range of elevated temperatures. It also showed moderate saline tolerance in aqueous media and bentonite-based drilling fluids and could therefore be regarded as a suitable drilling mud modifier. The polyelectrolyte provided excellent viscosity build-up efficiency and fluid-loss control (<15 cm³ /30 min) in bentonite and salt contaminated muds at temperatures between 120°C and 160°C. Its salt tolerance in aqueous monovalent and divalent electrolytic media indicates its potential usefulness in enhanced oil recovery.

Key words: polyelectrolyte; drilling fluid; rheological property; filtration loss; salt tolerance; temperature resistance.

1. Introduction

Drilling fluids are mixtures of natural and synthetic chemical compounds used to cool and lubricate the drill bit, clean the hole bottom, carry cuttings to the surface, control formation pressures, and improve the function of the drill string and tools in the hole [1], and assist in the collection and interpretation of information available from drilling cuttings, cores, and electrical logs [2]. In order to perform their fundamental functions, drilling fluids should possess several desirable characteristics, which greatly enhance the efficiency of the drilling operation [3]. Fluid properties can be highly influenced and altered by many factors in deep/ultra-deep drilling. Elevated temperature and pressure are among the most significant factors, in addition to ageing and electrolysis contamination. Elevated temperature might be introduced from geothermal source. Hydrostatic pressure, which is a function of depth, increases with drilling depth. Salts can be present in drilling fluids through fluids contamination during drilling of salt beds or purposely added as component during formulation of drilling fluids to obtain a salt saturated water based mud, or in offshore operations where seawater is used in preparing the drilling fluids [4]. Naturally, oil-based drilling fluids are more suitable in hostile environments owing to high performance as they are less sensitive to the borehole environment; provide high lubricity and deliver better drilling efficiency. However, environmental considerations regarding the fluid disposal and miscellaneous costs are factors which limit their applications [5-7]. Conversely, water based drilling fluids are more economical and ecofriendly although they are highly sensitive to formation characteristics, promote clay hydration and swelling which tend to increase well construction costs and additionally, less versatile for different wellbore conditions [8]. Water based drilling fluids are more commonly used compared to oil based fluids for both environmental and economic reasons [9].

Selection of the drilling fluid is based on the desired rheological parameters and filtration properties by the engineer keeping in view the borehole conditions [10]. A major difficulty in formulating high-temperature drilling muds is that the mud is negatively affected by elevated temperatures, first gelling excessively and then becoming inert at extreme temperatures [6]. For that reason, there is always a need for drilling mud additives which can stabilize drilling mud suspensions at high temperatures [11].

Filtration properties are regarded one of the very important characteristics of all drilling fluids [12]. One of the most critical functions of drilling fluids is to try to minimize the amount of drilling fluid filtrate entering the hydrocarbon bearing formation. The drilling fluid filtrates can cause formation damage owing to rock wettability changes, fines migration, clay swelling, drilling fluid solids plugging and formation water chemistry incompatibilities [7,13]. Moreover, this nature and thickness of the filter cake deposited on the borehole wall may cause differential pressure sticking [12].

Fluid loss control agents are additives used in drilling mud formulation to reduce the loss of fluid from the mud into the drilled formation. This fluid loss control help to maintain hole integrity, protect water sensitive shale, minimize hole washout to achieve better casing cement jobs, reduce fluid loss to productive formation, minimize formation damage and to reduce log analysis problems [14-15]. Over the years, some water soluble polymers have demonstrated effectiveness in reducing fluid losses in drilling fluids. The mechanisms of filtration control are based on polymer-clay electrostatic/entropic interaction and/or viscosifying properties of the polymers, resulting in reduction of filtration rate [16-17]. At moderate temperatures, polyacrylamide, biopolymers, starch, starch derivatives and cellulose derivatives provide fluid loss control. However, at elevated temperatures, these polymers tend to degrade thermally or oxidatively resulting in possible collapse of desirable rheological properties and filtration control with attendant poor performance or downright failure of the drilling fluid.

In an attempt to address part of the challenges associated with drilling fluids, the suitability of a synthetic polyelectrolyte as viscosifying agent and fluid loss reducer in water based drilling fluid aimed at elevated temperature and saline environment was examined.

2. Experimental Section

2.1 Synthesis of acrylamide-sodium acrylate-sodium 2-Acrylamido-2-methylpropane sulfonate -N, N'-dimethyl acrylamide- polyelectrolyte

All chemicals including mainly, sodium hydroxide, acrylamide (AM), acrylic acid (AA), N, N-dimethyl acrylamide (DMAM), sodium metabisulfite, ammonium persulfate and except 2-acrylamido-2-methylpropane sulfonic acid (AMPS) were of analytical grade and used without further purification.

The polyelectrolyte consisting of Acrylamide (AM), sodium salt of acrylic acid (AA), N,N-Dimethyl acrylamide (DMAM), 2-Acrylamido-2-methylpropane sulfonic acid (AMPS) polyelectrolyte was synthesised by free radical polymerization in aqueous sodium chloride medium. The reaction was conducted in a three-neck flask equipped with a magnetic stirrer, nitrogen gas inlet tube and a thermostatic water bath under nitrogen sparging using $(\text{NH}_4)_2\text{S}_2\text{O}_8/\text{Na}_2\text{S}_2\text{O}_5$ redox initiation. Reaction optimization conditions used were total monomer concentration of 20.38-31.28 wt/v, pH of 7.5-9.5, temperature of 40-60°C, initiator dosage of 0.4-0.8 wt%, and reaction time of 1.5-3.5 hours, using monomers feeds: AA of 20-28%, AM of 36-48%, DMAM of 8-12% and AMPS of 20-28%. AA and AMPS monomers were first dissolved separately in 0.5 molL^{-1} NaCl aqueous solutions and the pH adjusted to the basic range using 2 molL^{-1} NaOH. Acrylamide and was dissolved separately in 0.5 molL^{-1} NaCl aqueous solution whilst N,N-dimethyl acrylamide was mixed with the alkaline solution of AA before adding to the bulk. The pH of the mixture was finally adjusted to the desired basic medium value whilst ensuring a constant reactants system volume of 100ml. The reaction mixture was degassed of oxygen for at least 20 minutes prior to initiation of the polymerization. Reaction was terminated after the desired time. The polyelectrolyte was then purified using excess acetone.

2.2 Characterization of the Polyelectrolyte

The presence of the monomers was established by Fourier Transform-Infrared spectroscopy. The sample was finely ground to powder and analyzed using NICOLET 6700 FT-IR spectrometer

(Thermo Scientific SMART OMNI-SAMPLER, USA), recording the infrared spectra in the wave number range of $4,000\text{-}400\text{cm}^{-1}$

The intrinsic viscosity was determined using Brookfield DV-III Ultra programmable Rheometer (Brookfield Engineering Lab, Middleboro, USA) based on API procedure [18]. Different polymer concentrations were prepared in 3%NaCl and viscosity measured at 30rpm. Intrinsic viscosity was determined graphically using Huggins' and Kraemer's equations.

2.3 Properties Evaluation of Polyelectrolyte

2.3.1 Effect of Salinity and Hardness on Polyelectrolyte Viscosity

The apparent viscosities of polymer solutions were measured using Brookfield DV-III ultra rheometer (Brookfield Engineering Laboratories Inc., Middleboro, MA, USA) at shear rate of 100 and 200rpm. Polymer solutions were prepared in different concentrations of NaCl, CaCl_2 , MgCl_2 , and $\text{NaCl}:\text{CaCl}_2:\text{MgCl}_2(20:1:1)$. Salt tolerance was then evaluated by plots of polymer solution viscosities against salt concentrations as shown in Figs. 1-3.

2.3.2 Formulation and Test of Drilling Fluids

A stock base mud consisting of 500ml of tap water, 25g bentonite clay and 1.25g sodium carbonate per cup was prepared by mixing using electrically powered mechanical stirrer and allowed to age for at least 16 hours at ambient temperature. Test samples viz. Mud A, B, C, & D were then prepared by mixing the base mud and functional additives, depending on the formulation recipe, and blending to uniformity using a multi-speed mixer. Mud samples were tested initially and after 16hours of hot-rolled aging according to American Petroleum Institute(API) procedure [19] for measurement of rheological properties using ZNN-D6 six-speed rotary viscometer (Qingdao Haitonghai Specialized instrument Factory). API fluid loss was determined at pressure of 100psi using ZNS-A filter press tester. Test procedure after 16hr aging involved cooling the drilling fluid to room temperature, stirring in a mixing cup using the multi-mixer for 5min and conducting the tests. Tests results are shown in Tables 1-3 and Figs. 4-6.

3. Results and Discussion

3.1 Optimized synthesis of Polyelectrolyte

Optimum conditions for the synthesis of the polyelectrolyte was achieved at a mole ratio of AA :DMAM:AM:AMPS of 22:11:45:22, respectively using total monomer concentration of 22.95wt/v% and initiator dosage of 0.5wt% at reaction temperature of 45°C and pH of 8.0.

3.2 Characterization of the Polyelectrolyte

The intrinsic viscosity of the polyelectrolyte was obtained as 5.0dL/g.

Comparative analysis of FTIR spectra of polyelectrolytes with some known spectra of homopolymers and copolymers of the monomers confirmed the presence of the four monomers. Peak at 1662cm^{-1} was attributed to carbonyl stretching from AM, DMAM and AMPS. Carboxylate asymmetric stretching at 1573cm^{-1} and symmetric stretching at 1404cm^{-1} were notably assigned to AA. Peak at 1189cm^{-1} and 1044cm^{-1} were respectively, associated with asymmetric and symmetric stretching of sulfonate group of AMPS, amide characteristic peak of -CN- at 1452cm^{-1} was assigned to AM, DMAM and AMPS and rocking of C-H bond of methyl group at 1363cm^{-1} to DMAM and AMPS. The composition of the AM-AA-AMPS-DMAM polyelectrolyte was therefore validated.

3.3 Salinity Effect on Solution Viscosity of Polyelectrolyte

The high solubility of the polyelectrolyte in a wide range of aqueous media resulted from polyelectrolyticity based on carboxylate and sulfonate groups in addition to the polarity of the amide groups. Figs 1-3 present the effects of salinity and hardness on the viscosity of the polyelectrolyte in aqueous media. Initial decline in viscosity followed by relative stability were observed as concentrations of electrolyte increased. The initial viscosity loss was attributed to shielding effect of ionic groups, which reduces repulsion and causes polymer chain folding in aqueous brine solution. However, the viscosity enhancing potential of the polyelectrolyte is attributable to its anionic groups which repel each other causing the polymer chains to adopt a

more expanded and rigid-rod-like conformation resulting in increase in hydrodynamic volume, in addition to polymeric amide-water hydrogen bonding. Observably, figure 3 illustrates anti-aging stability of the polyelectrolyte at 80°C in 11.0% brine solution (NaCl/CaCl₂/MgCl₂) for the period of 10days tested.

3.4 Effect of Temperature

3.4.1 Rheological Behaviour

Figures 4 -5 and Table 1 show the relative stability of the polymer-incorporated mud at elevated temperatures. Apparent viscosity of Mud A before and after hot-aging was fairly high and stable with average value of about 50cP up to a temperature of 150°C but dropped by almost 36 % (to 32cP) at 160°C. Demonstrating relatively higher thermal stability of polyelectrolyte as mud additive compared to KPAM with a viscosity loss of 81% under the same conditions. Corresponding plastic viscosity of the increased to a peak value at 150°C and declined at 160°C whilst the yield point decreased gradually from 28cP before aging to 6cP after hot-aging at 160°C. The blank completely lost its yield point and gel strength after hot-aging at 120°C whilst KPAM based fluid showed a yield point of unity and zero gel strength after hot-aging at 160°C.

Table 1 Thermal Effect of Mud Properties

Mud A	Blank	PE/KPAM								
Water	500ml	500ml								
Bentonite clay	25g	25g								
Na ₂ CO ₃	1.25g	1.25g								
Polymer	-	4.0g								
			Drilling mud @ ambient temp.		Drilling mud after hot-rolled aging					
					HR @ 120°C		HR @ 140°C	HR @ 150°C	HR @ 160°C	
Parameter	Blank	PE	KPAM	Blank	PE	PE	PE	PE	KPAM	PE
Apparent Viscosity (AV), cP	11.3	52	52.5	10	49	50.5	50	10	32	
Plastic Viscosity (PV), cP	3.5	24	30	10	34	41	42	9	26	
Yield point (YP), cP	7.8	28	22.5	0	15	9.5	8	1	6	
10 Sec gel (G10''), cP	7	9.5	6.5	0	2	1	1	0	0.5	
10 min gel (G10'), cP	9.5	19.5	11	0	3	1.5	1.5	0	0.75	
API Filtrate loss, ml/30min	24	10	NC	107	10.5	10	9.5	11	9.0	

3.4.2 Fluid loss Control

Blank mud filtration loss before dynamic hot aging was 24ml//30min. However, after aging at 120°C it exhibited excessively high fluid loss of 107ml/30min. The introduction of the polyelectrolyte in the mud effectively reduced and maintained filtration loss within 9.0-10.5ml/30min before and after hot-aging between 120°C-160°C as shown in fig 5. Whilst the polyelectrolyte offered thermal stability in the polymer-incorporated mud, the blank suffered severe degradation, flocculation or dehydration in the absence of the polyelectrolyte.

3.5 Salinity Effect at Elevated Temperature

Rheological Properties

Tables 2 - 3 and fig. 6 present the salinity effect of the mud at elevated temperatures. In Table 2, the apparent viscosity and plastic viscosity, respectively, increased from 77.5cP to 85cP and 15cP to 42cP while yield point decreased from 62.5cP to 43cP, in the presence of 5% NaCl, after hot-aging at 120°C. Corresponding KPAM mud values changed from 84cP to

86cP, 16cP to 64cP with about 67% loss of yield point (67cP to 22cP) after hot-aging. Similar rheological behaviour was observed in Table 3 with slight modification of mud composition and aging temperature at 150°C. In mud D, presented on fig. 6, the addition of 0.8% poly-electrolyte concentration in the mud resulted in nearly seven-fold increase in fluid viscosity in the presence of 5% NaCl, 0.25% CaCl₂, and 0.25% MgCl₂. Indicating that the monovalent and divalent ions would have slight influence on the performance of salt contaminated mud when fortified with the polyelectrolyte.

Table 2 Saline Effect on Mud Properties at 120°C

Mud B	Blank	PE/KPAM				
Water	500ml	500ml				
Bentonite clay	25g	25g				
Na ₂ CO ₃	1.25g	1.25g				
SPNH	7.5g	7.5g				
Lignite	7.5g	7.5g				
NaCl	25g	25g				
Barite	100g	100g				
Polymer	-	4.0g				
	Drilling mud @ ambient temp			Drilling mud hot-rolled @ 120°C for 16 hr		
Parameter	Blank	PE	KPAM	Blank	PE	KPAM
Apparent Viscosity (AV), cP	16.5	77.5	84	13	85	86
Plastic Viscosity (PV), cP	9	15	16	6	42	64
Yield point (YP), cP	7.5	62.5	67	7	43	22
10 Sec gel (G10''), cP	8.5	49	55.5	5	21	15
10 min gel (G10'), cP	10	52.5	57.5	5.5	22.5	20
API Filtrate loss, ml/30min	42	6.0	6.5	71.0	6.5	7.5

Table 3 Saline Effect of Mud Properties at 150°C

Mud C	Blank	PE/KPAM/XCD						
Water	500ml	500ml						
Bentonite clay	25g	25g						
Na ₂ CO ₃	1.25g	1.25g						
SPNH	7.5g	7.5g						
Lignite	7.5g	7.5g						
NaCl	25g	25g						
Barite	100g	100g						
Polymer	-	3.0g						
	Drilling mud @ ambient temp				Drilling mud hot-rolled @ 150°C for 16 hr			
Parameter	Blank	PE	KPAM	XCD	Blank	PE	KPAM	XCD
Apparent Viscosity (AV), cP	16.5	66	77.5	75	12	62	54	55
Plastic Viscosity (PV), cP	7	15	10	13	11	51	42	29
Yield point (YP), cP	9.5	51	66.5	62	1	11	12	26
10 Sec gel (G10'')	10	37.5	48	33.0	0.5	3.5	6	8.5
10 min gel (G10')	8.5	43	45	33.5	1.0	4.0	7	10
API Filtrate loss, ml/30min	42	6.7	6.0	6.0	82.0	12.0	11.0	15.0

Filtration Control

In Table 2, whilst the blank lost its filtration control from 42.0 to 71.0 ml/30min, revealing weak viscosity retention potential at high temperature, the polyelectrolyte-incorporated mud maintained better filtration control (6.0 to 6.5 ml/30min) and rheological properties and therefore exhibited superior anti-aging performance than both KPAM and the blank.

Mud C as indicated in Table 3 had similar composition to Mud B except for slight reduction in polymer concentration and increase in aging temperature. As expected, the presence of polyelectrolyte, KPAM or xanthan biopolymer demonstrated considerable improvement in rheological properties and filtration control compared to the blank. Whilst KPAM apparently showed higher effectiveness in reduction of filtration loss, the polyelectrolyte manifested fairly higher dual-functional efficiency in filtration control and viscosity retention after aging at 150°C.

In Mud D, the polyelectrolyte solely effected and maintained a much lower filtration loss of 4.5ml/30min before and after 120°C hot-aging compared to blank filtrate volume of 82ml/30min. In the blank mud, the high chemical contamination coupled with high temperature caused dehydration and flocculation of the bentonite, resulting in a much higher filtration rate, in spite of the higher solid content.

3.5.1 Mechanism and Properties of Polyelectrolyte in Drilling Fluid

Enhancement of rheological properties of Mud A, B, C & D following the addition of polyelectrolyte is attributed to the anionic character, water solubility and structural hydrophilicity of the polyelectrolyte. The anionic groups repel each other possibly via double layer forces, which cause the polymer chain to adopt a more expanded, rigid-rod-like conformation, in addition to the clay-polymer electrostatic interaction and polymeric amide-water hydrogen bonding. This result in a large hydrodynamic volume and consequently enhancement of rheological properties of the muds. The relatively high viscosity build-up efficiency of the polyelectrolyte suggests that the drill mud would offer sufficient resistance to reduce slip velocity of the cuttings and effectively suspend cuttings during drilling operations.

The main chain or backbone of a polymer plays a key role in thermal stability. Structurally, the incorporation of the large side group AMPS together with acrylamide-based dimethyl group into the polyelectrolyte effectively improved its thermal and hydrolytic resistance and thus enhanced its superior rheological and filtration control functionality at elevated temperatures as compared to KPAM, xantham biopolymer and the blank.

Filtration losses in all the mud systems before and after heat treatment between 120°C-150°C were acceptably low. The mechanism of filtration control is effected through polymer-clay adsorption and viscosifying potential of the polymer which reduce water mobility through the clay pores. The polyelectrolyte functions as a protective colloid with the solids in the mud, forming a thin low permeable filter cake which drastically reduces the amount of fluid permeating into the pore spaces. This would potentially maintain hole integrity, protect water sensitive shales, minimize formation damage and reduce log analysis problems.

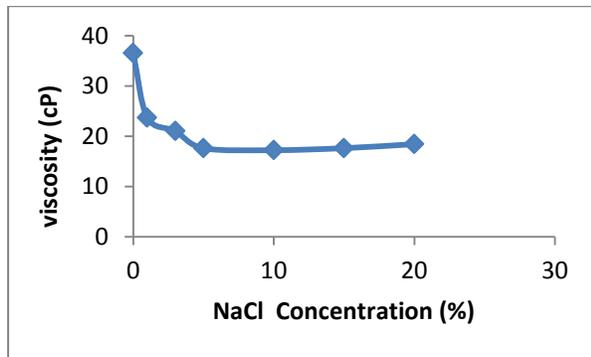


Fig. 1 Effect of NaCl on the viscosity of polyelectrolyte at shear rate of 200rpm at 40°C

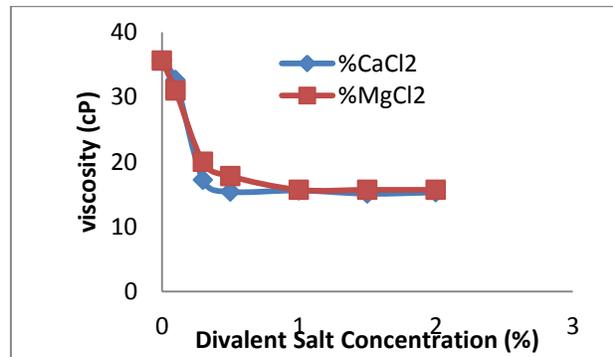


Fig. 2 Effect of divalent salt on the viscosity of polyelectrolyte at shear rate of 200rpm at 40°C

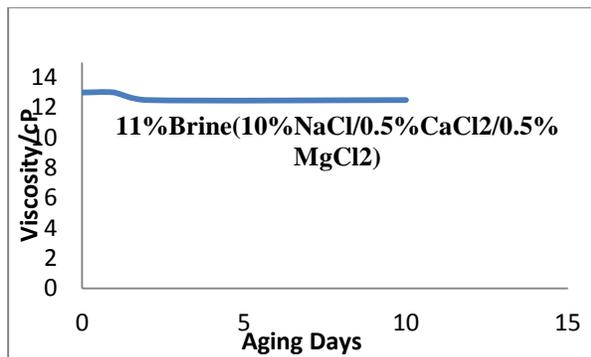


Fig. 3 Effect of aging time at 80°C on polyelectrolyte in 110,000ppm brine at shear rate of 100rpm

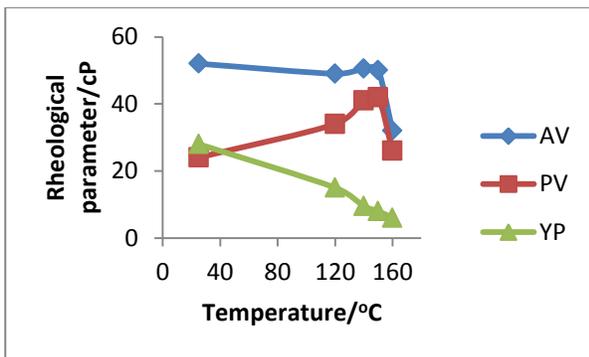


Fig. 4 Thermal Effect on Rheological Properties of Mud with Polyelectrolyte Additive

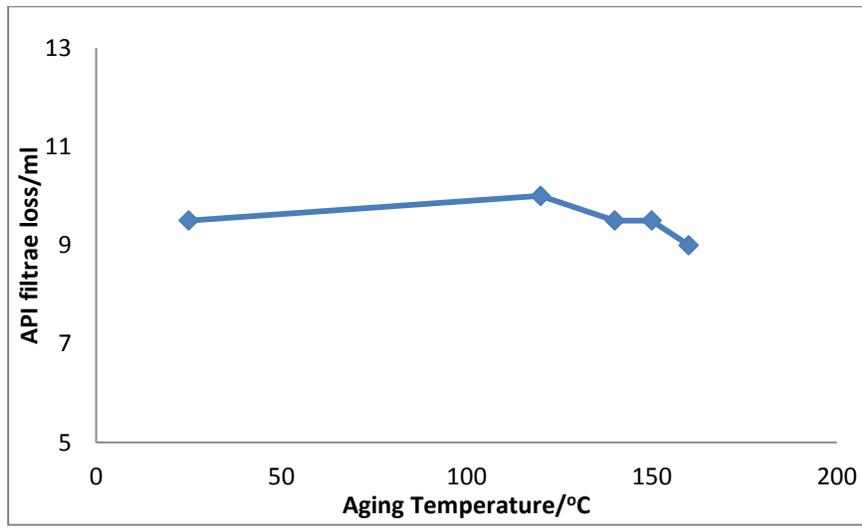


Fig. 5 Thermal Effect on Fluid Loss of Mud with Polyelectrolyte Additive

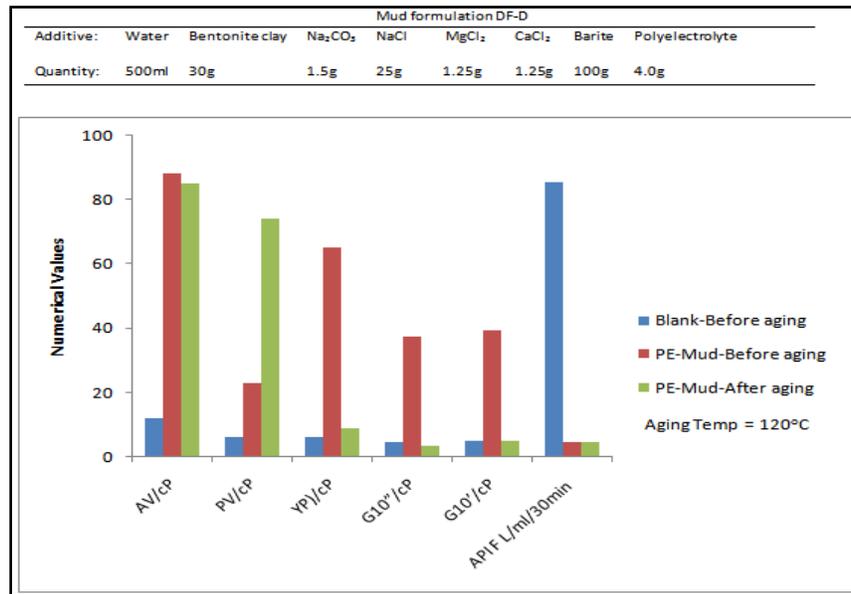


Fig. 6 Effect of salt contamination on mud properties

4. Conclusion

The polyelectrolyte has potential usefulness and versatility in salt-free and salt contaminated drilling fluid systems. It is an excellent fluid-loss reducer in bentonite-based drilling muds. It also exhibited high viscosity enhancing properties. The polyelectrolyte could be used as a stabilizer for bentonite-based drilling muds by providing good temperature resistance and anti-aging performance. It would be a suitable additive in water-based drilling mud for application in relatively high saline environment possibly above 160°C.

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Abbreviations and Formulae

AMPS	2-Acrylamido-2-methylpropane sulfonic acid	AV	apparent viscosity
DMAM	N, N-dimethyl acrylamide	PV	plastic viscosity
AA	acrylic acid	YP	yield point
AM	Acrylamide	$G_{10''}$	gel strength at 10sec
PE	Polyelectrolyte	$G_{10'}$	gel strength at 10min
KPAM	Potassium polyacrylamide/acrylate	FL	filtrate loss
XCD	Xantham biopolymer	API	American Petroleum Institute
HPAM	partially hydrolyzed polyacrylamide	AV	$\frac{1}{2}\Phi_{600}$
SPNH	sulfomethyl humate and phenolic resin	PV	$\Phi_{600} - \Phi_{300}$
Φ_{600}	Dial reading at 600rpm	YP	AV-PV
Φ_{300}	Dial reading at 300rpm	G_{10}	$\frac{1}{2}\Phi_3$
Φ_3	Maximum dial reading at 3rpm		

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