





Article

Optimization of Partially Hydrolyzed Polyacrylamide (HPAM) Utilized in Water-Based Mud While Drilling

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Abstract: Water-soluble polymers are becoming increasingly important in various applications, such as stabilizer fluids and drilling muds. These materials are used as viscosifiers and filtration control agents, flocculants, and deflocculants due to their superior properties in increasing viscosity and gelling ability in the presence of crosslinkers. In general, studying the rheological behavior of drilling fluids is of paramount importance to ensure successful well drilling operations. Partially hydrolyzed polyacrylamide is one of the polymers widely used in water-based muds. The main objective of this study is to optimize the rheological properties of drilling muds through a characterization study of various parameters, including rheological behavior, viscosity, temperature (23 °C, 40 °C, and 60 °C), salinity using KCl and NaCl contents, aging, pH, solubility, and structural analysis using infrared of partially hydrolyzed polyacrylamide. The study aims to demonstrate the importance of using polymers in drilling muds. The findings revealed that a rate of 3% of HPAM gave better rheological behavior, the influence of KCl (1.5%, 3%, and 4.5%) was greater than that of NaCl (1.5%, 3%, and 4.5%) on polymers, and the aging test showed that the different formulations are stable and maintain their behavior up to 110 °C. The solubility test results confirmed the maximum amount absorbed by polyacrylamide ($[C_{HPAM}] = 66.42$ g/L) in order to avoid aggregation, gelification, and enhance the drilling mud by utilizing the prescribed contents.

Keywords: HPAM; rheology; thermal aging; aging; optimization; drilling mud

1. Introduction

Currently, water-soluble polymers are very important for many industrial applications [1,2]. The oil industry, in particular, is a major consumer of polymers for various oil recovery operations, such as drilling, fracking, modification of permeability, cementation, clogging, and control of the mobility of the aqueous phases [3,4]. The purpose of using polymers is to avoid serious problems during drilling such as wellbore stability, which is the foundation and prerequisite for efficient methane production from offshore gas hydrates. Nonetheless, as a result of the invasion and disturbance caused by drilling mud, gas hydrates around the wellbore gradually dissociate, resulting in a rapid reduction in sediment strength and, as a result, borehole collapse [5]. The use of polymers in drilling offers a number of environmental benefits such as: (1) the protection of the geological formations during drilling by reducing subsurface formation disruption and preventing gas and liquid leaks; (2) reducing drilling downtime by lowering difficulties with drilling fluid viscosity, temperature, and pressure; and (3) increasing viscosity to reduce the amount of water required for drilling, which not only aids in water conservation but also reduces the adsorption of CO₂ thickeners in shallow shale soil particles, serving as one way to protect the shale environment and water cycle [6].

Polymer flooding is a chemical method that is widely used in enhanced oil recovery (EOR). It involves changing the rheological properties of the displacement fluid by adding water soluble polymers [7–10]. A wide range of polymers, such as carboxymethylcellulose (CMC), xanthan gum, starches, and polyanionic cellulose (PAC), have been used in drilling fluid engineering to improve drilling fluid properties and avoid problems during drilling operations [11–15]. In addition, eco-friendly additives such as starch, polyphenols, and lignosulfonate showed a good filtration performance in Bentonite-water-based drilling fluids [16], along with the use of single-walled carbon nanotubes/polyvinyl pyrrolidone (SWCNs/PVP) nanocomposites on the drilling efficiency of water-based muds, with the last forming a protective layer on the shale surface to obstruct the access of water and offering structural stability of the drilling mud [17]. Partially hydrolyzed polyacrylamide (HPAM) is a water-soluble polymer material used in oil fields [18,19]. It usually has a linear structure with high molecular weight [20] and is obtained by partial hydrolysis of PAM or by copolymerization of sodium acrylate with acrylamide [21]. Furthermore, HPAM is traditionally used with KCl as a polymer, which acts by encapsulation of clay [22], at very high concentration, for the coating of the clays, improving the load capacity of the overburden and functioning as power lubricant mud.

In areas where there are problems of stability of wells, the polymers are usually filled with salt providing a cation to help stabilize the training; the most common polymer used to replace oil mud is mud HPAM/KCl system [23]. It has the advantages of improving the stability of clays and having good load capacity. Its drawbacks are due to contamination by solids and reduced thermal stability [20,24]. Besides, the HPAM plays the role of viscosifiant, friction reducer, and flocculant when it brings a reduction of the filtrate [18]. It can be used in drilling fluids containing small amounts of solids that have those groaning, in either freshwater or saltwater [25,26], in sealing the microcracks and covering the surfaces of clay with a film that delays the dispersion and disintegration. In addition, KCl is used as only a clay inhibitor in most muds of HPAM-designed and has strong ability to form hydrogen bonding, which is helpful for swelling, but not for dissolving. Many studies in the literature discussed the use of HPAM in the field of oil industry, e.g., Zhang and Fang found that with purification of polymer, extensive treatment of polymer solutions, molecular weight (Mw), radius of gyration (Rg), and hydrodynamic radius (Rh) of these polymers can be well characterized by light scattering [27]; Pereira et al. demonstrated that the successful usage of bentonite and organic bentonite clays as reinforcement fillers in HPAM/PEI-based systems [28].

In terms of infectivity, these materials had no effect on the viscosity of the solutions, which is a critical factor in operational issues [28]. Kuma et al. investigated modified CMC and PHPA polymer drilling fluids with NaCl and KCl and haematite mineral where

KCl and NaCl acted as great swelling inhibitors for drilling muds, while haematite just slightly increased the rheological properties on CMC–bentonite mud and did not have much effect on the PHPA–bentonite mud [26]. Das et al. compared CMC and HPAM and it was found that the PHPA mud exhibited a higher effective viscosity, apparent viscosity, and plastic viscosity than the CMC mud; the PHPA also showed higher values of yield point and gel strength which indicated that PHPA mud has a better cutting-carrying capacity than CMC mud [19]. Laura et al. compared partially hydrolyzed polyacrylamide and xanthan gum (XG). As it was expected, HPAM solutions and nanopolymer sols were more sensitive to salinity and temperature changes than the XG sols [29]. Oliveira et al. concluded that the elemental analysis enabled the tracking of the evolution of the HPAM's hydrolysis degree at various temperatures over 360 days under reservoir conditions [8]. It is important to note, however, that the use of polymers in the drilling industry must be carefully managed in order to avoid environmental contamination. Polymers must be disposed of properly, and the number of polymers used must be monitored to minimize any negative environmental effects.

This article reports on the formulation of two types of salt NaCl and KCl at different concentrations where the HPAM was applied as a polymer in these experiments. Their characteristic relationships were observed, established, and compared as the main focus of this article. In particular, we studied the effect of KCl and NaCl with various concentrations present in the solution of HPAM in order to characterize the effect of salts on rheological properties and to select the capability of adding the polymer on water-based mud at different condition of concentration, temperature, pH, and time for improved oil recovery in reservoir conditions.

2. Materials and Methods

2.1. Devices

The following devices were used throughout the research work, Model 900 viscometer, Modular Compact Rheometer MCR 302e, electronic balance, pH/ORP Meter, FTIR spectrometer, and oven.

2.2. Chemicals

The used chemicals include partially hydrolyzed polyacrylamide (HPAM) with high molar mass (MW) of an average of 20×10^6 (Da), and a hydrolyze degree of 30%; A (1%), A' (1.2%), A1 (2%), and A2 (3%), sodium chloride (NaCl), N1 (1.5%), N2 (2.5%), and N3 (4.5%), potassium chloride (KCl), K1 (1.5%), K2 (2.5%), and K3 (4.5%), and distilled water (1000 mL).

2.3. Experiment Procedure

Preparation of 4 solutions of HPAM with different concentration (0.6%, 1.2%, 2%, and 3%); 1 solution was selected HPAM (3%); and temperature varied between 23 °C and 60 °C. Then, amounts of NaCl and KCl were added to prepare three solutions for each, and then the pH was measured. The solutions were agitated magnetically. The aging test consists of aging the polymer HPAM under controlled conditions of temperature and time. The test was carried out using an oven for 1 week with a temperature increase from 60 to 110 °C. Infrared test and solubility test were also carried out (see Figure 1).

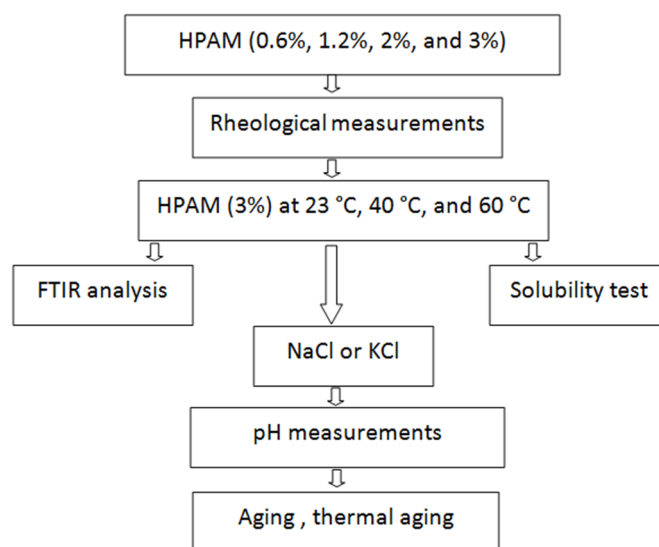


Figure 1. Methodology flow chart.

2.4. Measurements

Rheology Measurement

The rheological behavior of the four solutions was determined by OFITE model 900 Viscometer, which is a true Couette coaxial cylinder rotational viscometer with 12 fixed speeds (600, 300, 200, 100, 60, 30, 20, 10, 6, 3, 2, and 1), and a heat cup made of stainless steel with power (150 Watts) and a maximum recommended temp of 190 °F (88 °C). For a fully automated Control/Data Acquisition System suitable for research applications, the Model 900 Viscometer may be connected to a computer via a serial (RS-232) port using OFITE's exclusive and field-proven Windows™-based ORCAD[®] software [30]. While, the rheological behavior of the solutions (A2 + NaCl, A2 + KCl, and aged solutions) were determined by Modular Compact Rheometer: MCR 302e with cone plate (CP) geometry, this rheometer is characterized with its stable results even for long-term measurements with low torque, a trimming mirror for a 360° view of the sample with no blind spots, which avoids sample preparation errors and leads to highly reproducible results, and a fast one-hand coupling and decoupling of measuring geometries in just one second—even at extreme temperatures (−160 °C to 1000 °C): novel quick-connect mechanism [31].

The solutions HPAM passed in a viscometer where the RPM varied (10, 60, 100, 200, 300, 600), and the solutions A2, A2+ NaCl and A2+ KCl passed in a rheometer to measure viscosity, shear rate, and shear stress.

2.5. Infrared Spectroscopy

Infrared spectroscopy (FTIR spectrometer vertex 70, RAM II module) was used to measure the FTIR spectra of HPAM sample. The data were collected at a spectral resolution of 4 cm^{−1} within the scanning range of 4000–400 cm^{−1}.

3. Results and Discussions

3.1. FTIR Analysis

The interpretation of the different bands of the spectrum obtained during the analysis by FTIR spectrometry of partially hydrolyzed polyacrylamide without any modification are based on the absorption peaks (or bands, then presented in transmission diagrams). Furthermore, in terms of wave numbers, we can extract the types of chemical bonds and functional groups present in the molecules, and use spectrum tables to identify the different characteristic bands of this spectrum (Table 1). The HMPA polymer was analyzed using FTIR analysis, and the results are displayed in Figure 2. The peak at 1632 and 3230 cm^{−1} represent the stretching vibrations of the C-O and N-H groups of the −CONH₂ group,

respectively. These peaks confirm that the acrylamide group is present in the synthesized polymer. The absorption bands at 2963 and 2925 cm^{-1} indicate the stretching vibrations of the $-\text{CH}_3$ group, while the absorption bands at 2853 and 1457 cm^{-1} correspond to the $-\text{CH}_2-$ bonding in the polymer chain, and absorption bands at 1116 cm^{-1} indicate the presence of $-\text{CN}$ stretching. The infrared spectrum of A2 solution is illustrated in Figure 1.

Table 1. Main infrared bands observed in solution A2.

Wave Numbers (cm^{-1})	The Corresponding Link
2800–2950	C-H
3500–3400	O-H
3230	N-H
900–1200	C-N
1647	C=O

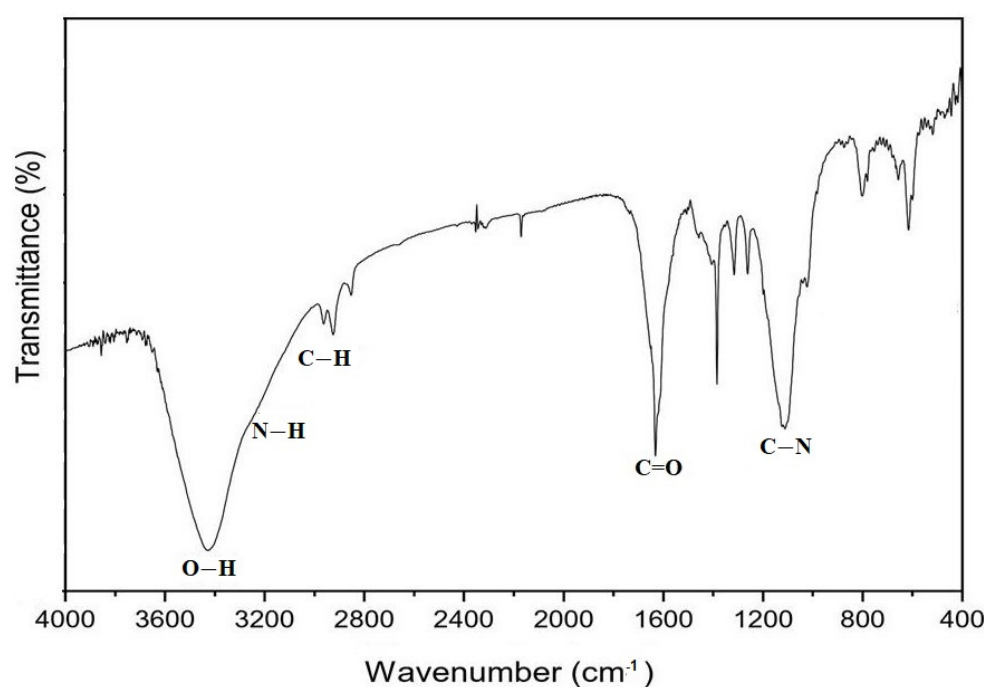


Figure 2. IR spectrum of solution A2.

3.2. Effect of the Concentration and Heat on the Rheological Behavior of HPAM Polymer Concentration

The rheology of a fluid refers to its flow behavior, which can be affected by various factors including the concentration of hydrolyzed polyacrylamide (HPAM) present in the fluid. HPAM is a commonly used thickener in various industrial processes such as oil and gas drilling, mining, and wastewater treatment.

The effect mechanism of HPAM concentration on rheology can be explained by its impact on the polymer chain entanglement and the formation of a three-dimensional network structure. At low HPAM concentrations, the polymer chains are not sufficiently entangled, resulting in a low viscosity fluid. As the concentration of HPAM is increased, the polymer chains become more entangled, which leads to an increase in the viscosity of the fluid. This is because the polymer chains form a three-dimensional network structure that impedes the flow of the fluid, making it more resistant to deformation.

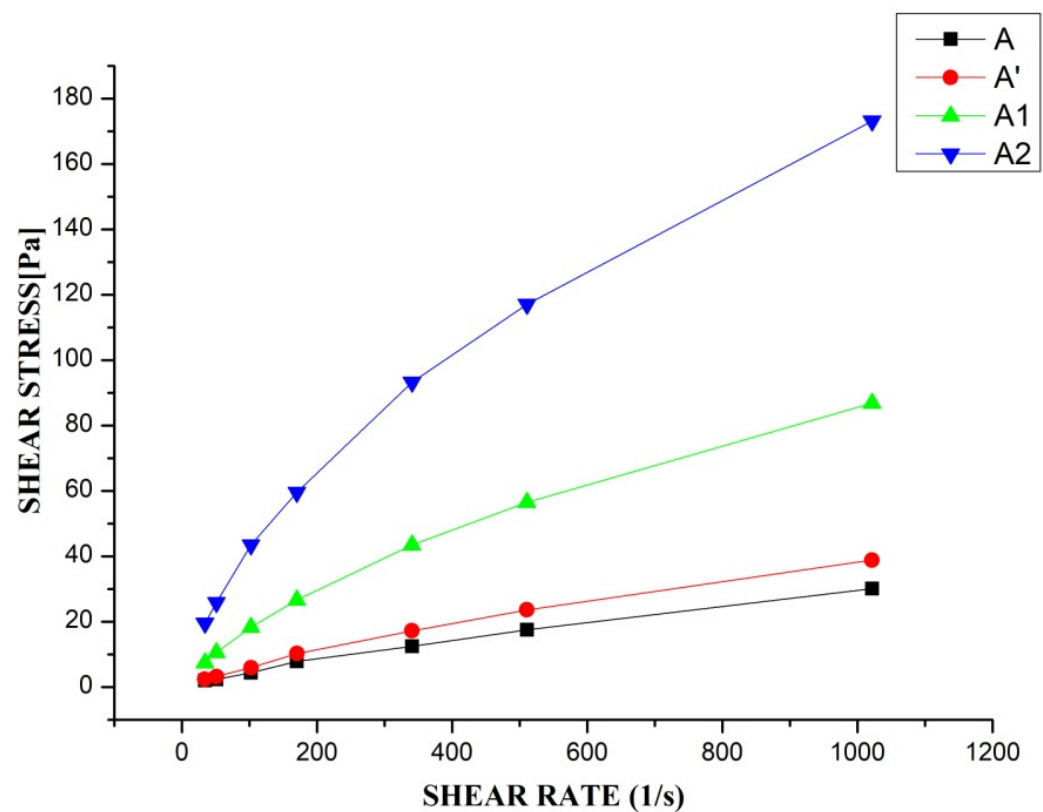
Furthermore, the molecular weight of HPAM also plays a role in its effect on rheology. At higher molecular weights, HPAM molecules are more entangled, resulting in a more pronounced thickening effect. Additionally, the degree of hydrolysis of the HPAM also affects its rheology. As the degree of hydrolysis increases, the polymer chains become shorter, leading to a decrease in viscosity.

In summary, the effect of HPAM concentration on rheology can be attributed to its impact on the polymer chain entanglement and the formation of a three-dimensional network structure.

Drilling fluid rheological behavior can be simulated using various empirical rheological models, yielding several meaningful model parameters and thus providing a more direct physical interpretation of their complicated flow behavior [32,33].

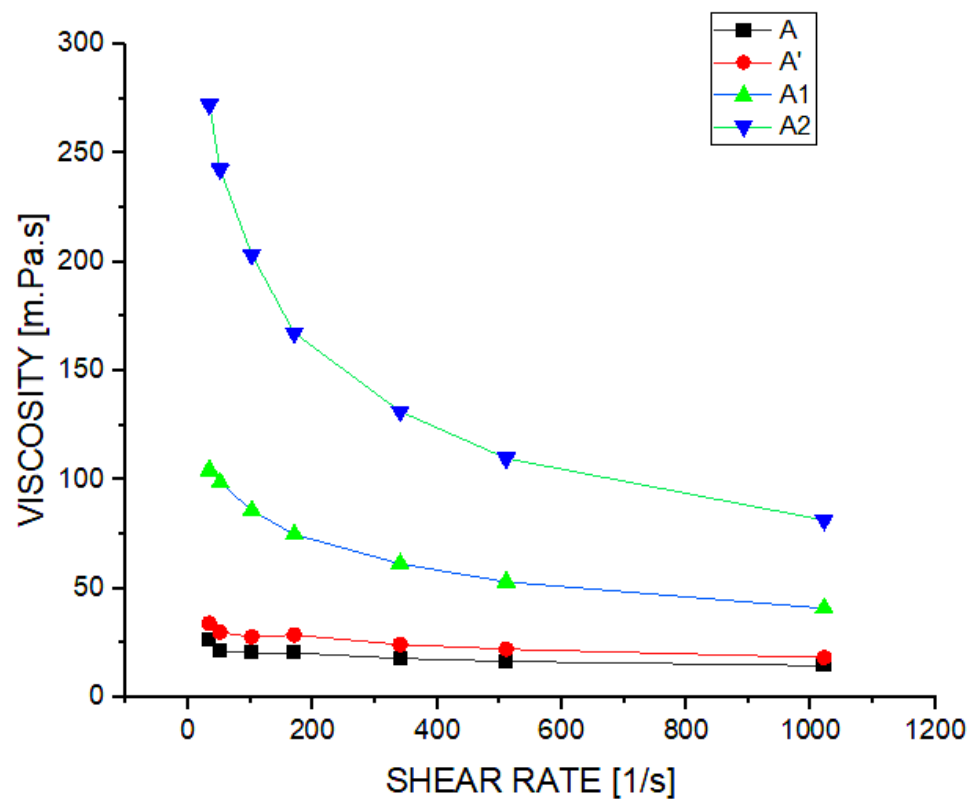
The rheological behavior of various suspensions is illustrated by the variation of the shear stress as a function of the shear rate under varying additive concentrations of HPAM (1%, 1.2%, 2%, and 3%). From the rheograms in Figure 3a, all suspensions show a non-Newtonian flow. The rheological properties of HPAM suspensions depend primarily on the degree of association of the molecules and therefore on the polymer concentration regime in which we are placed. The molecular weight is a parameter that largely influences the rheological results [27,34–36].

Adsorption and flocculation are the results of hydrogen bonds between the solid-liquid surfaces and the hydroxyl groups of the polymers; hence, the variation of the viscosity of the aqueous dispersions of HPAM, at different concentrations, was examined as a function of the shear rate, and the decrease in viscosity as a function of the shear rate, shown in Figure 3b, is explained by the fact that carboxyl groups in the HPAM molecules in collaboration with water molecules, which provides the dispersion resulting in increased friction between particles. It is observed that A2 gave a better result. Compared to the literature, three common empirical models are used to simulate shear stress curves versus shear rate: Bingham-Plastic, power law, and Herschel-Bulkley [32]. The Herschel-Bulkley model is the appropriate one to describe the rheological properties of suspensions as HPAM behaves in solution as a pseudoplastic fluid with threshold stress [33,37,38].



(a)

Figure 3. Cont.



(b)

Figure 3. (a) Rheograms of HPAM suspensions at different concentration, (b) viscosities of HPAM suspensions at different concentration as a function of shear rate.

3.3. Heat

Figure 4a,b, respectively, present the variation of the stress and the viscosity depending on the shear rate of solution A2 at different temperatures (23 °C, 40 °C, and 60 °C).

It has been found that a decrease in the shear stress during an increase in temperature is due to the movement of molecules (increasing thermal agitation and thus increasing the speed of travel).

The thermo-dependent pseudo-plastic model takes into account the variation in viscosity depending on the shear rate and temperature. An increase in temperature results in a decrease in viscosity due to the increase in mobility of the polymer chains so that the increase of temperature would still break down intermolecular hydrophobic associations unavoidably, and hydrolyse the acrylamide group in HPAM into acrylate groups [18].

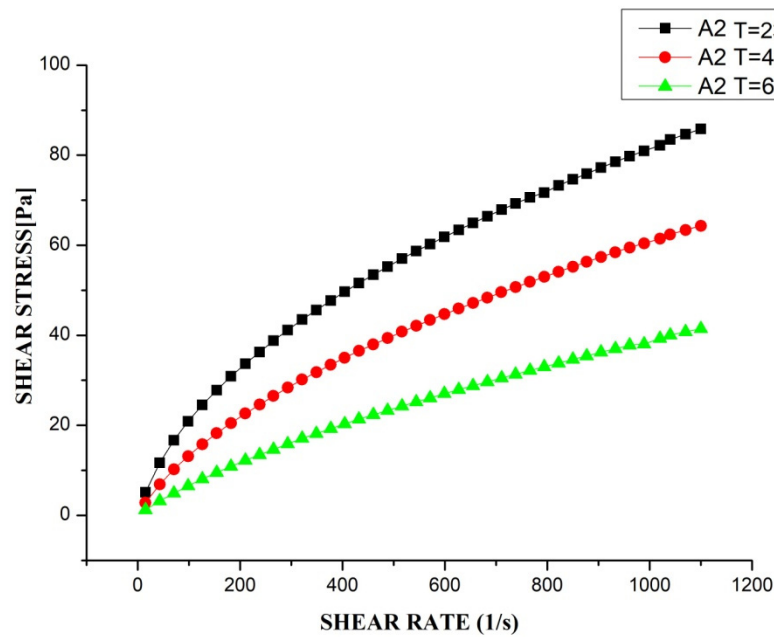
3.4. Effect of Concentrations of NaCl and KCl and Heat

3.4.1. NaCl

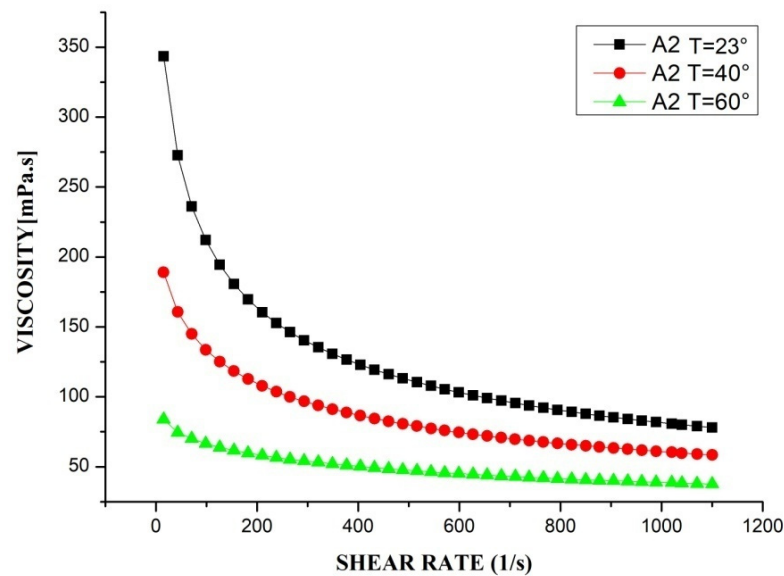
Figure 5a,b present, respectively, the variation of the stress and the viscosity as a function of the shear rate of A2 solution modified by NaCl at different temperatures.

It can be noted, however, that the same change is seen in the case of NaCl but with a lower efficiency.

The ambient temperature increase within the range of 40 °C and 60 °C will decrease the shear stress (increasing the temperature promotes intra-molecular movement). By addition of salt (electrostatic effects examination), the behavior of polyelectrolyte joins the neutral polymer. In the absence of added salt, the ionic force is low and the electrostatic effects are important. This ionic strength increases as the concentration of added salt increases and the examination/screening effects greatly reduce the electrostatic interaction [39].



(a)



(b)

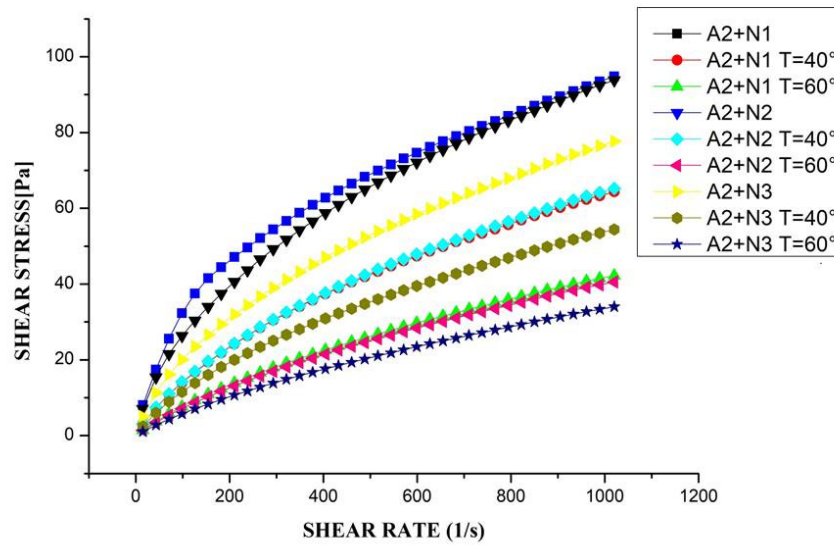
Figure 4. (a) Rheograms of HPAM suspensions at different temperatures, (b) viscosities of HPAM suspensions at different temperatures as a function of shear rate.

3.4.2. KCl

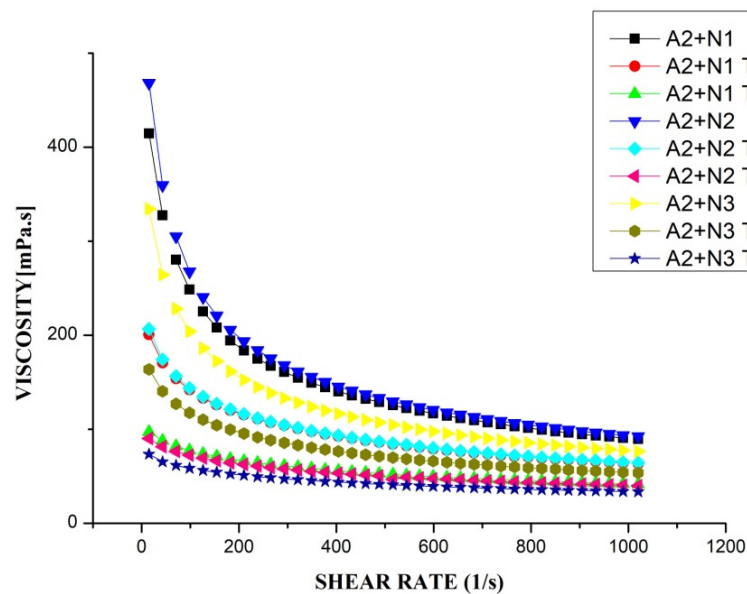
Figure 6a,b present, respectively, the variation of the stress and the viscosity as a function of the shear rate of A2 solution modified by KCl at different temperatures.

The increase in the amount of salt used results in a decrease in the shear stress which is explained by the efficiency of the KCl in this mixture, i.e., high solubility of KCl and small decrease compared to NaCl which is less effective.

Increasing the ambient temperature within the range from 40 °C to 60 °C will decrease the shear stress (increasing the temperature promotes intra-molecular movement).



(a)



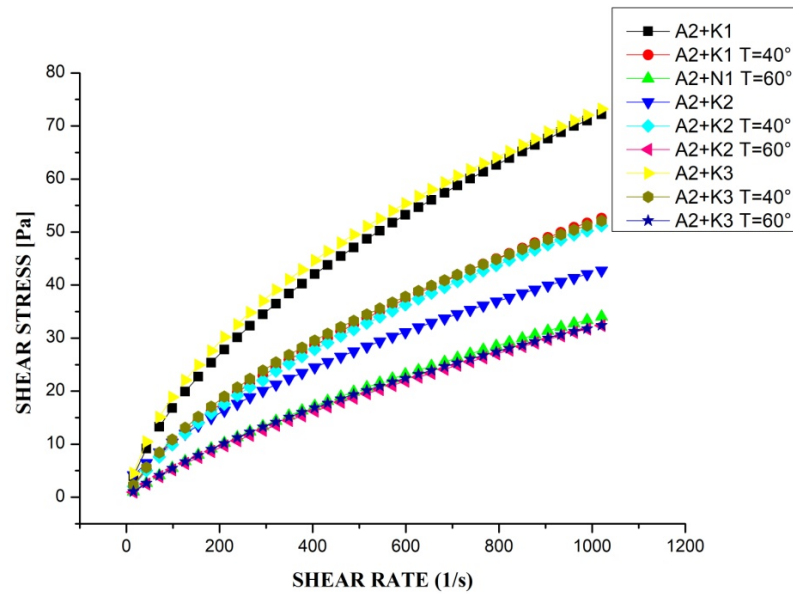
(b)

Figure 5. (a) Rheograms of HPAM suspensions at different concentration of NaCl and temperatures, (b) viscosities of HPAM suspensions at different concentration of NaCl and temperatures as a function of shear rate.

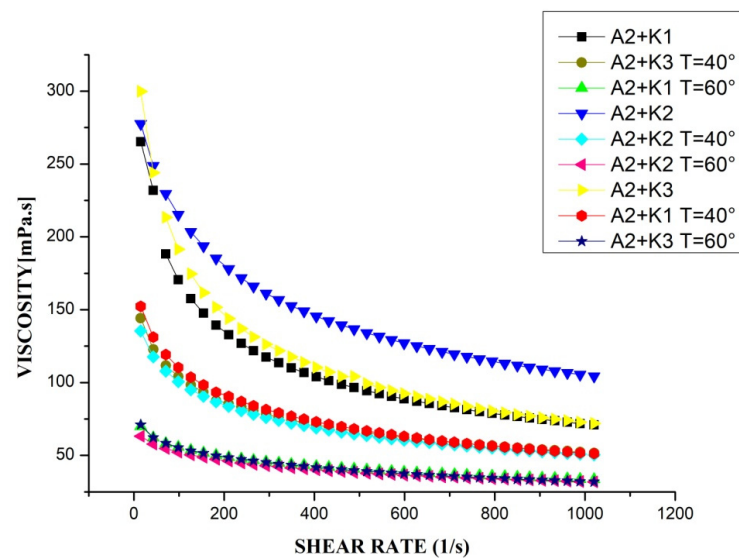
The very important dependence of viscosity on temperature is well illustrated by the flow curves. This is the case for the vast majority of fluids such as blood, shampoos, high molecular weight polymer solutions, and others. Concerning polymer solutions, the interpretation of the decrease in viscosity is explained by the changes in the molecular structures of polymer solutions for which the fluid becomes less viscous [40].

The increase in the quantity of salt (KCl) decreases the viscosity (decrease in the threshold of viscosity), meaning adding salt will make the mixture less viscous.

The influence of NaCl is less important than that of KCl, which is explained by the lower solubility of NaCl and also that our treated polymer contains sodium Na in its composition, minimizing its effectiveness.



(a)



(b)

Figure 6. (a) Rheograms of HPAM suspensions at different concentration of KCl and temperatures, (b) viscosities of HPAM suspensions at different concentration of KCl and temperatures as a function of shear rate.

3.5. Effect of the Aging Time on the pH of Different HPAM Suspensions

Aging is a slow and irreversible evolution of one or more material properties from a reference point, usually at the end of the manufacturing [41]. This development may be the result of changes in the macromolecules that ensure their mechanical cohesion, their composition, or its morphology [42].

pH is a chemical property of the mud that is often related to its stability and also to its ability to easily disperse the clay materials from which it load during drilling [43]. The pH determination provides the free ions concentration H^+ (or OH^-). Drilling sludge is generally basic and its pH may vary from 8 to 10 depending on the case [44].

Figure 7a–c, respectively, show the variation in pH as a function of time of polyacrylamide formulations diluted in water and modified with KCl and NaCl.

It was found that:

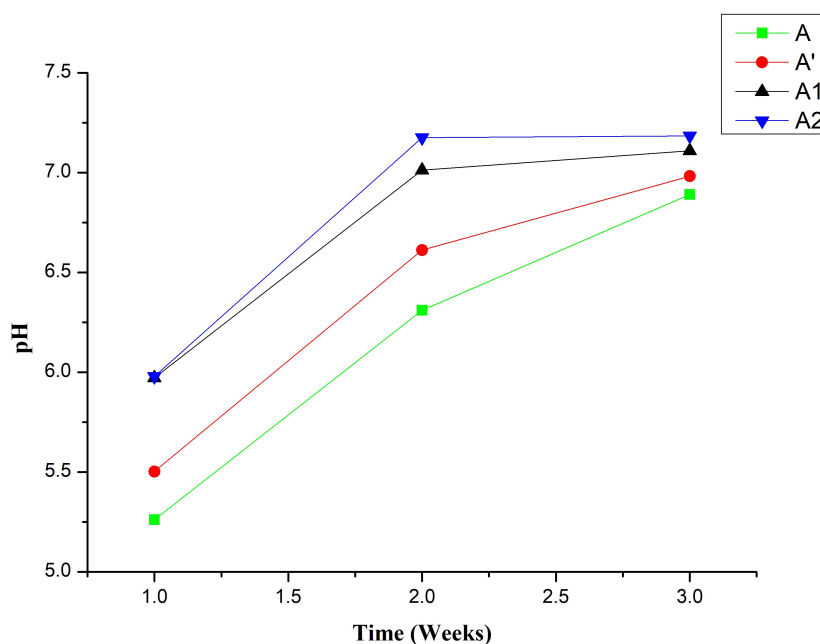
- (1) The concentration of the polymer in the solution affects the pH of the water. With increasing concentration, the pH increases but afterwards becomes almost constant;
- (2) The variation in KCl concentration affects the pH of the solution but after 48 h the solutions stabilize;
- (3) The influence of NaCl on the solution is less important compared to KCl because of the presence of the Na⁺ and Cl⁻ ions; additionally, the effect of KCl on rheology is typically more significant than sodium chloride (NaCl) due to differences in their ionic size and hydration properties.

3.6. Thermal Aging

The HPAM solution was aged thermally in order to follow its aging status. Figure 8 shows the texture of the solution as a function of time and heat. Table 2 summarizes the status and the colors of each phase.

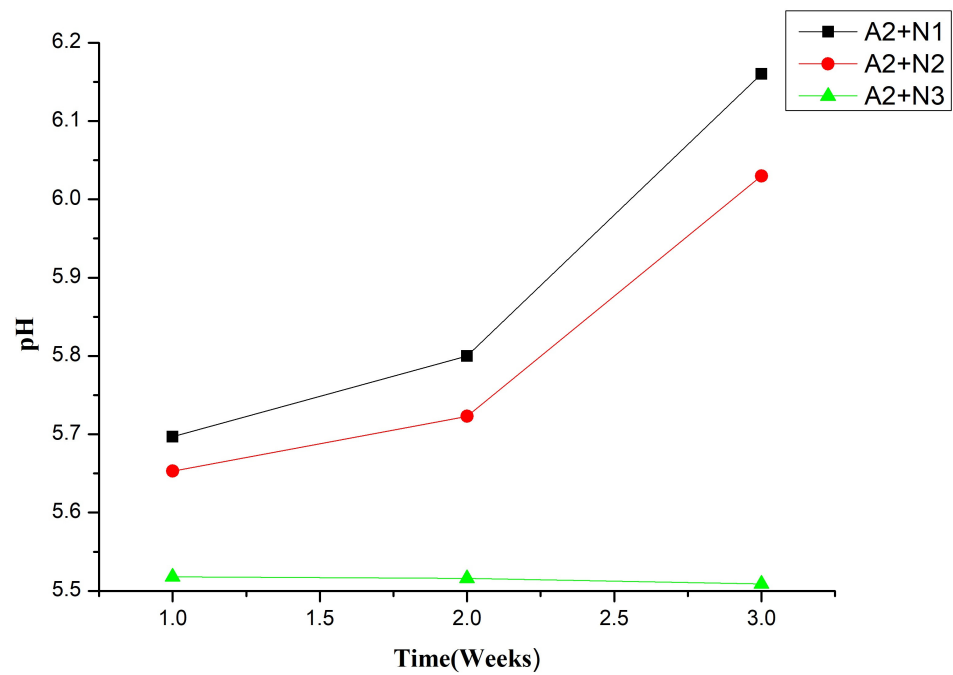
Table 2. HPAM solutions color and status before and after aging.

Solution	Color and Status
(a)	Transparent solution.
(b)	No changes have been noted.
(c)	HPAM solution color changed to ward yellow because of the temperature, Polymer precipitation and the volume decreased a bit.
(d)	The HPAM's color has changed to ward yellow and the volume has become lower.
(e)	No changes had been noted except the volume decreased.
(f)	The solution was vaporized completely and the color had become darker.
(g)	The solution was vaporized and turn into white powder.

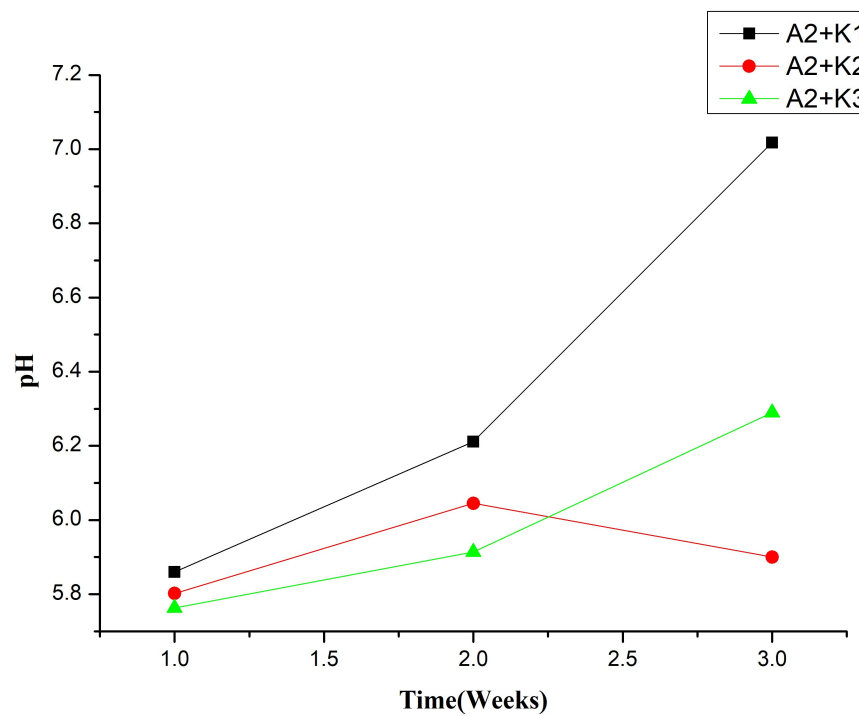


(a)

Figure 7. Cont.



(b)



(c)

Figure 7. (a) pH as a function of time for HPAM solutions, (b) pH as a function of time for HPAM solutions + NaCl, (c) pH as a function of time for HPAM solutions + KCl.

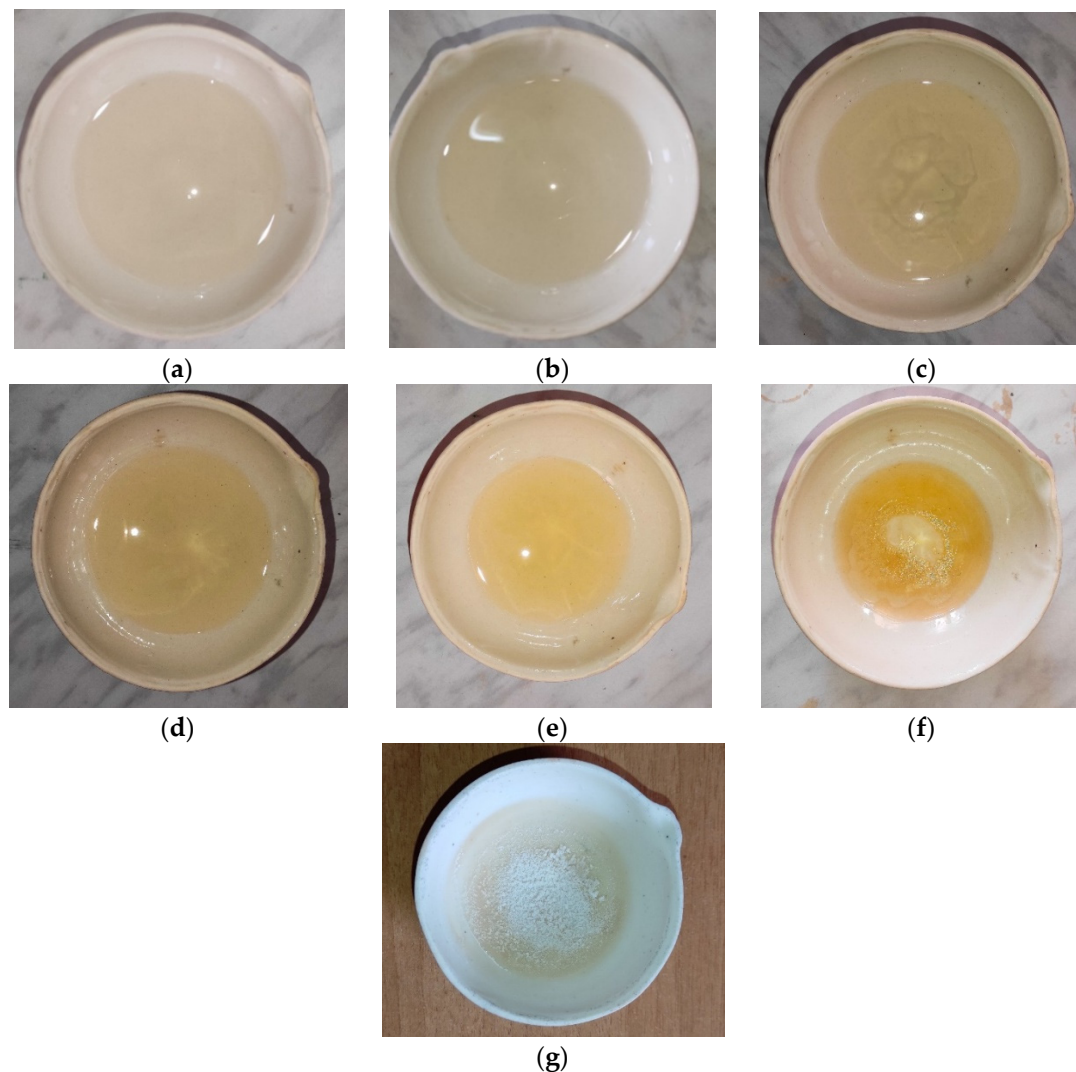


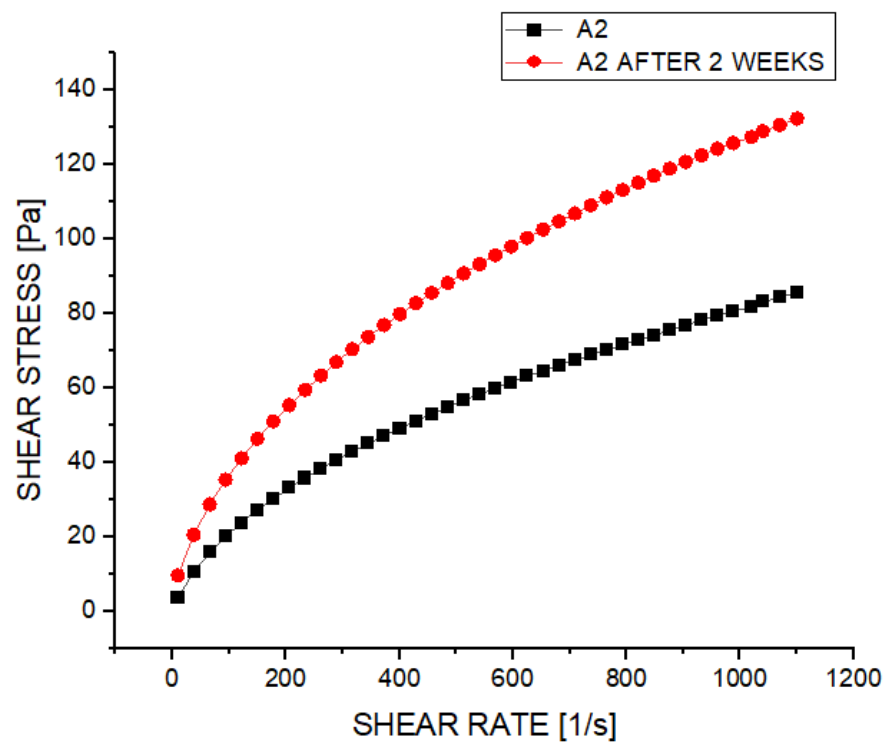
Figure 8. (a) (A2) solution before aging test, (b) after 24 h in the oven at 60 °C, (c) after 48 h, 24 h in the oven at 90 °C, (d) after 96 h, 48 h in the oven at 110°, (e) after 144 h, 96 h in the oven at 110°, (f) after 192 h, 144 h in the oven at 110°, (g) after completely 8 days in the oven at 110°.

3.7. Aging Time

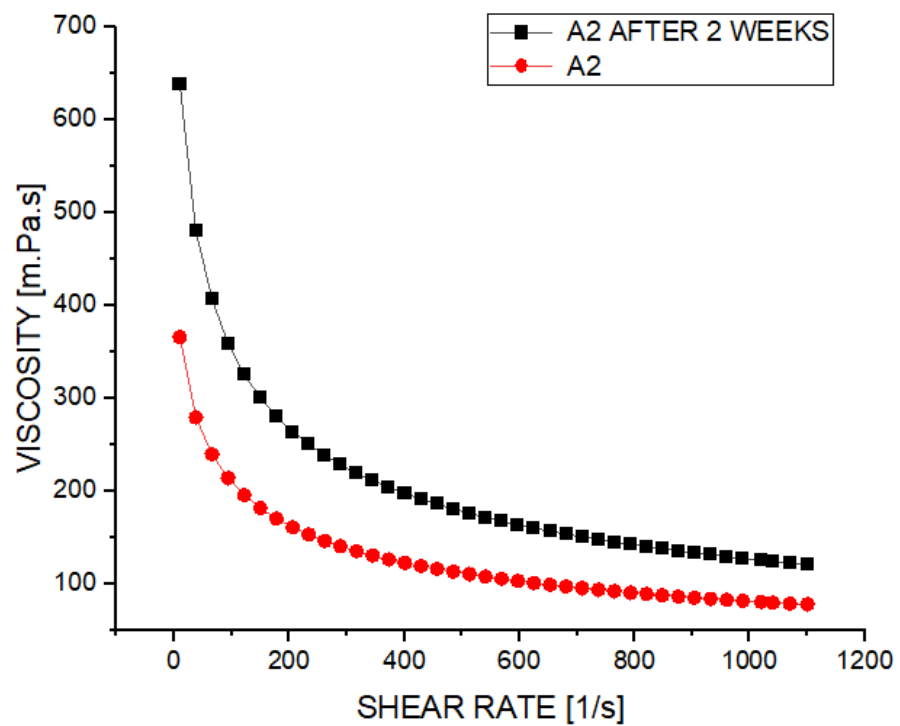
Figures 9–11 present, respectively, the variation of the stress and the viscosity depending on the shear rate of the 2 weeks aged A2, and 1 week aged A2 + K1, A2 + K2, A2 + K3, A2 + N1, A2 + N2, and A2 + N3.

It has been noted that an increase in the viscosity is due to the cross-linked feature of polyacrylamide. Cross-linker significantly increases the viscosity of linear gel by increasing the molecular weight of the base polymer by linking multiple molecules together. Cross-linker increases molecular weight without additional polymers.

Cross-linked variants of polyacrylamide have shown greater resistance to degradation. Hence, they are more stable for longer periods. Therefore, linking of polymer chains through chemical linkages gives material a more rigid structure and potentially a better-defined shape [45].

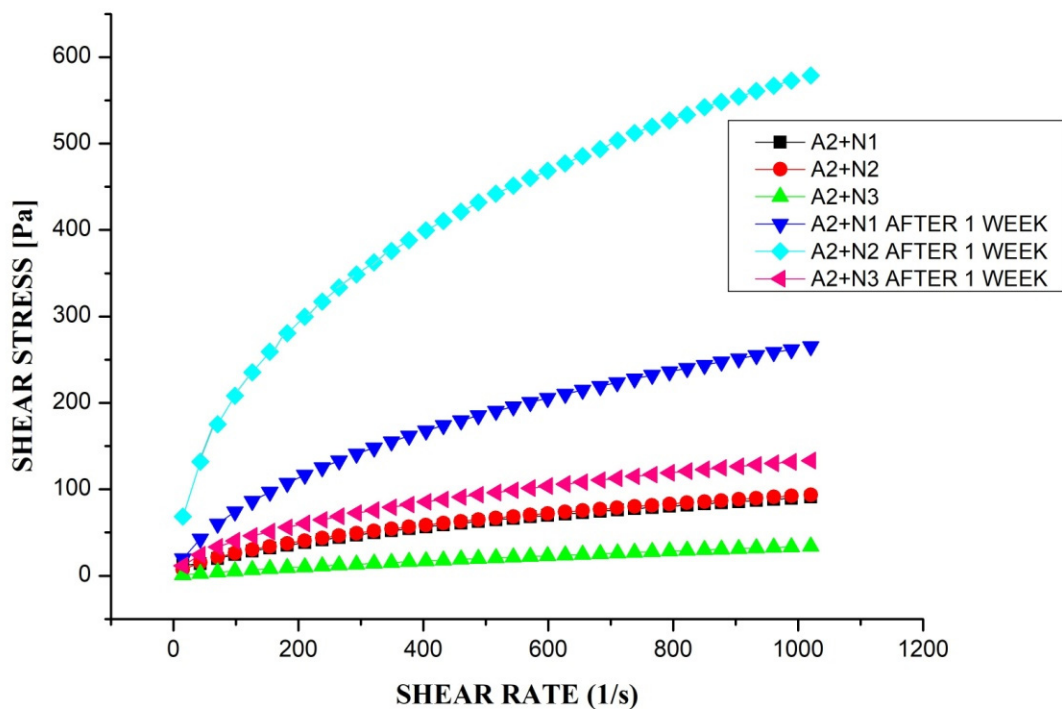


(a)

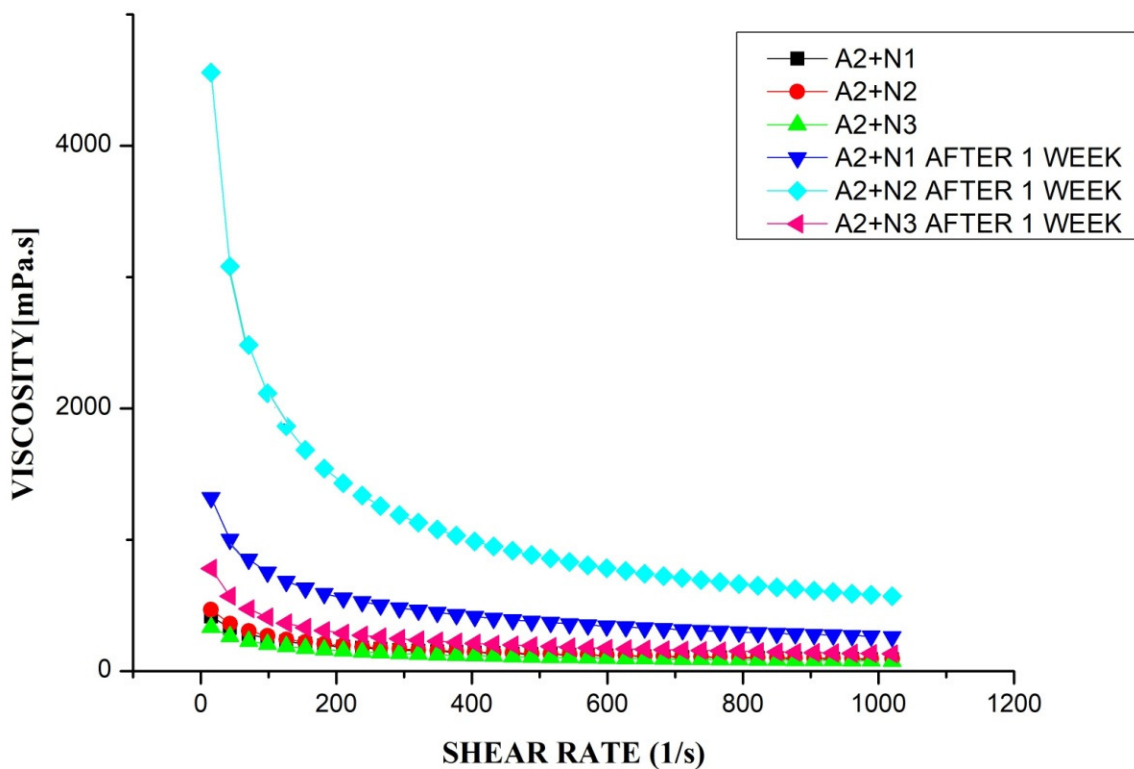


(b)

Figure 9. (a) Shear stress of A2 solution as a function of shear rate after and before 2 weeks, (b) viscosity as a function of shear rate after and before 2 weeks.

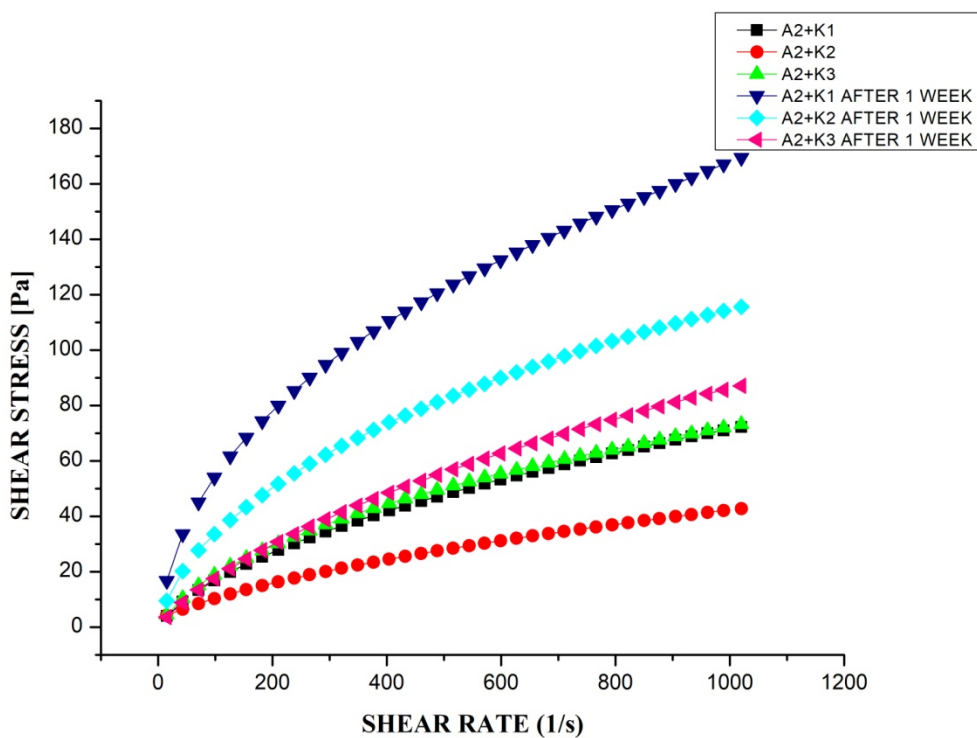


(a)

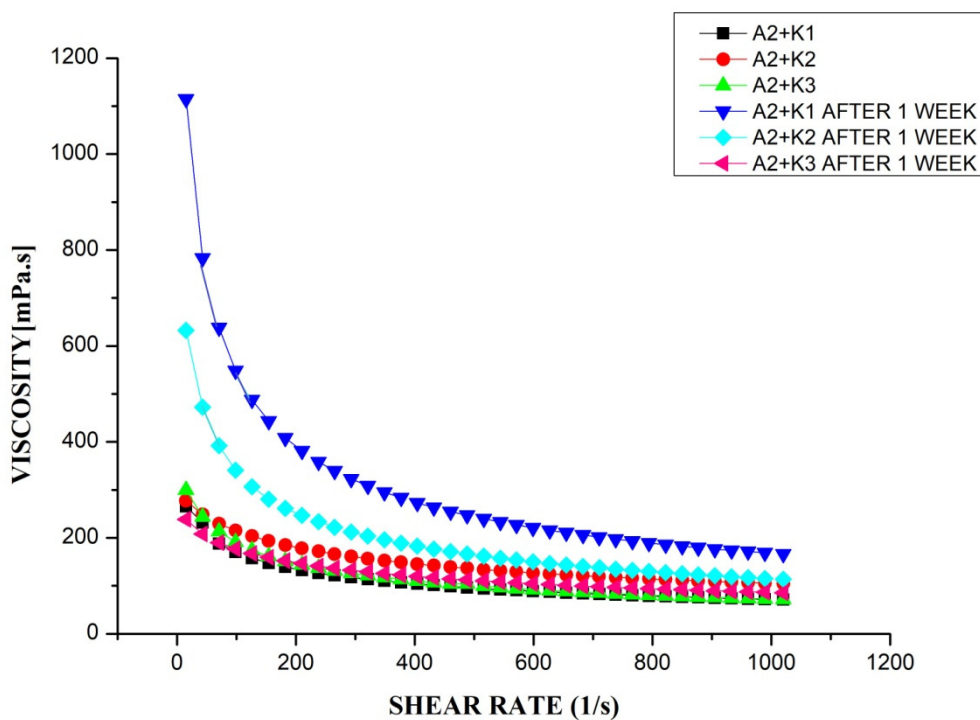


(b)

Figure 10. (a) Shear stress of A2 solution as a function of shear rate at different NaCl concentrations before and after 1 week, (b) viscosity as a function of shear rate.



(a)



(b)

Figure 11. (a) Shear stress of A2 solution as a function of shear rate at different KCl concentrations before and after 1 week, (b) viscosity as a function of shear rate.

3.8. Solubility Test

Solubility is described as the maximum amount of substance that can dissolve in a certain volume of water [46,47]. For a given temperature, solubility will depend on the structure of the compound and the nature of the solvent. Impurities can considerably affect the solubility of a substance in water [25,48].

In our experiment, we took 40 mL of the solvent (distilled water) and added 1 g of the polymer each time we see a total homogeneity (total dissolution of the polymer) (see Figure 12). Furthermore, we have to mix the solution for 15 min after each addition of polymer, until a saturated solution is obtained (it can no longer dissolve solute), and saturation is at: $[C_{\text{HPAM}}] = 66.42 \text{ g/L}$.

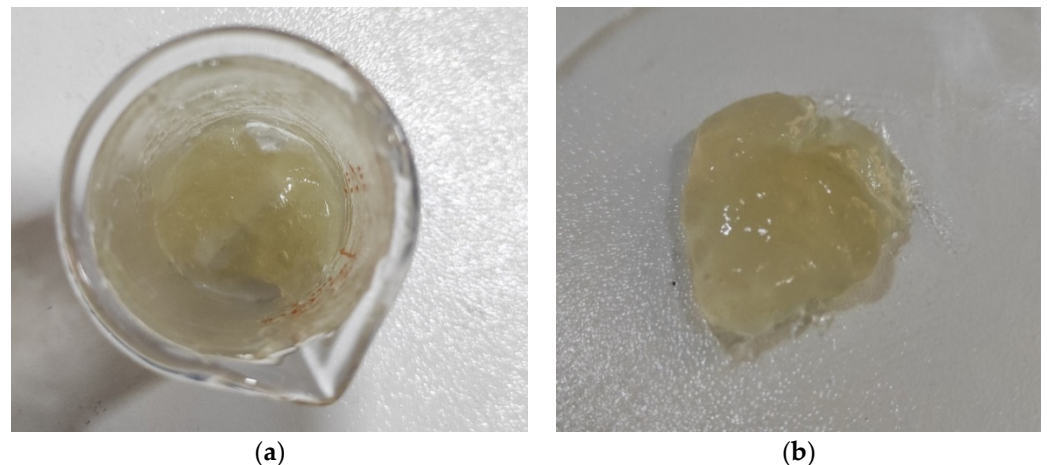


Figure 12. (a) 1 h from adding HPAM, (b) HPAM solubility.

4. Conclusions

This work presents experimental results on the rheological characteristics of partially hydrolyzed polyacrylamide (HPAM) in aqueous solutions with varying experimental conditions such as polymer concentration (0.6%, 1.2%, 2%, and 3%), temperature (20 °C, 40 °C, and 60 °C), salts, shear rate applied, and time. The molecular mass is a parameter that significantly affects the rheological results, and in this study, 3% of HPAM was selected. Temperature is another factor that affects the rheological properties, where the higher the temperature, the lower the rheological properties. Increasing shear rate, temperature, and salt concentration (KCl and NaCl) decrease the viscosity of the polymers. The influence of KCl is greater than that of NaCl. The aging test demonstrated that the different HPAM formulations are stable and maintain their behavior up to 110 °C. The solubility test results confirmed the maximum amount of partially hydrolyzed polyacrylamide that can be absorbed to avoid aggregation and gelification.

Various characterization techniques, including rheology, infrared spectrometer, aging test, and solubility test, were used to gather information on the properties of the new formulations prepared. The rheological tests confirmed the pseudoplastic behavior of all formulations and the effect of incorporating salts and bentonite on the behavior and reaction of HPAM under different conditions. The results demonstrated the capability of using HPAM under various conditions in water-based mud.

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