Available online www.jsaer.com

Journal of Scientific and Engineering Research, 2023, 10(11):145-155

Research Article

ISSN: 2394-2630
CODEN(USA): JSERBR

Experimental Evaluation of Polymer and Nanoparticle Hybrid for Enhanced Oil Recovery

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Abstract Enhanced oil recovery (EOR) techniques are important for increasing oil production as to meet global energy demands. Polymer flooding is a commonly used EOR method, but it has issues with polymer retention and vulnerability to degradation. Hybrid of polymers and nanoparticles reduces polymer retention, resists degradation, and maintains superior rheological properties. The study compares the impact of a bare polyacrylamide (PAM) polymer solution and a polyacrylamide-alumina nanoparticle (PAM/Al_2O_3 NP) hybrid in terms of viscosity, pH, efflux time, and oil recovery. Rheological measurements and sand pack flooding tests were conducted to determine the effects of the solutions. The mutual correlation between oil recovery and various parameters were generated using statistical model. The results indicate that the hybrid solution showed higher oil recovery efficiency (83.81%) compared to the bare PAM solution (72.22%) at a concentration of 0.3wt%. The hybrid solution also exhibited higher viscosity, efflux time, and pH (57.15cP, 1543sec, and 7.9, respectively) compared to the bare PAM solution (49.68cP, 1342sec, and 6.1, respectively) at the same concentration. Therefore, the addition of Al_2O_3 NP to the PAM solution modifies its properties, reduces PAM adsorption, preserves polymer chains, and maintains entanglement. This enhances the efficiency of the EOR process by ensuring stable and optimized fluid flow.

Keywords Enhanced Oil Recovery, polyacrylamide, nanoparticle, polymer nanohybrid, viscosity.

1. Introduction

The increasing global demand for energy has prompted a heightened emphasis on the extraction of crude oil from oil fields. Traditional primary and secondary recovery methods have only been able to retrieve approximately 50% of the original oil in place (OOIP), leaving a substantial volume of oil underground. Consequently, there is a need for a tertiary recovery method, known as Enhanced Oil Recovery (EOR), which has the potential to recover between 50-80% of the oil, depending on the type of crude oil and reservoir. This addresses the disparity between the escalating global energy demand and the insufficient recoveries from conventional methods [1, 2, 3]. Various EOR techniques, such as chemical, thermal, miscible, and microbial flooding, are being globally investigated to tackle these challenges and extract residual oil. Among these methods, chemical flooding, particularly polymer flooding, has demonstrated potential in recovering an additional 30-60% of the Original Oil in Place (OOIP), contingent on the reservoir and crude oil type [4].

Polymer flooding, a chemical method utilized in enhanced oil recovery, involves the introduction of polymers to brine to mitigate the difference in viscosity between the injected fluid and the reservoir fluids [5]. Among the various chemical techniques, polymer flooding is widely recognized as the predominant and efficacious approach for the prospective retrieval of oil and gas resources [6, 7]. Synthetic partially hydrolyzed polyacrylamide (PAM) and the biological polysaccharide xanthan are the most employed polymers for enhanced oil recovery. Xanthan gum, a biopolymer with high molecular weight and rigid polymer chains, exhibits thermal stability in the range of 70 °C to 90 °C, but degrades with increasing temperature and is susceptible to bacterial degradation [8, 9, 10]. Conversely, Polyacrylamide (PAM) is a synthetic, water-soluble polymer commonly utilized for polymer flooding due to its cost-effectiveness, viscosifying properties, well-established physiochemical characteristics, and effective mobility control. The thickening ability of PAM is attributed to its high molecular weight and the electrostatic repulsion between its polymer chains and segments [11]. PAM offers advantages such as resilience to mechanical forces during reservoir flooding, affordability, and resistance to bacterial degradation. It can be employed at temperatures up to 99 °C, contingent on the hardness of the brine [8]. Furthermore, the incorporation of nanoparticles in polymer nanohybrids has been investigated for potential enhancements in the rheological behavior of polymer solutions in chemical flooding operations. The reduced size and increased surface area of nanoparticles (NPs) make them suitable for use in polymer flooding [12]. Incorporating nanoparticles into the solution can also enhance the network structure of polyacrylamide (PAM) solution, resulting in improved mechanical and thermal properties [13].

Hu et al. (2021) examined the combined application of nanoparticles and polymer in enhanced oil recovery processes. They conducted water, polymer, and nanoparticle (Silica nanoparticle (SiO_2) and Alumina nanoparticle (Al_2O_3)) injections to assess the oil recovery factor. Their research indicated a decrease in retention and capillary forces, as well as an increase in polymer viscoelastic behavior, leading to enhanced oil recovery due to the favorable mobility ratio of polymer nanoparticles in fluid loss [14]. Maghzi et al. (2014) conducted an experiment demonstrating that the introduction of SiO_2 NPs into the PAM solution increased its viscosity, resulting in a 10% increase in oil recovery efficiency during a polymer flooding process [15].

This study aims to assess the application of a polymer, such as polyacrylamide (PAM), with alumina nanoparticles (Al_2O_3 NP), referred to as polymer nanohybrid (C_3H_5NO-Al_2O_3), to enhance the viscosity, pH, and efflux time of the PAM solution, thereby modifying oil viscosity and significantly improving oil recovery. The objectives of this investigation are as follows: to evaluate the impact of the polymer nanohybrid at various concentrations on viscosity, pH, efflux time, and oil recovery efficiency; and to validate the performance of the polymer nanohybrid (C_3H_5NO-Al_2O_3) and the PAM solution through mutual correlation. Despite the widespread use of simple polymer flooding as an enhanced oil recovery (EOR) method over the years, polymer susceptibility to shear and thermal degradation has been a concern [4]. As many reservoirs requiring EOR have high temperatures, polyacrylamide (PAM) solutions experience a significant decrease in viscosity and reduced oil recovery efficiency [11]. The need to address this challenge has led to the utilization of PAM/Al_2O_3 hybrid for enhanced oil recovery in the context of this study.

2. Materials and Methods

2.1 Materials

The materials and apparatus used to conduct experiment in this work includes, Crude oil sample, Polyacrylamide (PAM), Alumina nanoparticle (Al_2O_3 NP), Sodium chloride (NaCl), Potassium chloride (KCl), Prepared laboratory brine (NaCl + KCl), Encapsulated plug sample (unconsolidated formation), Set-up for flooding/liquid permeameter flow loop, U-tube viscometer, pH meter, Stop watch, Measuring cylinder and Magnetic stirrer.

2.2 Methods

2.2.1 Preparation of polymer solution and alumina nanohybrid

Polymer solutions were prepared by combining PAM (provided by Eddy Chemicals, Mile 1 Diobu, Port Harcourt, Rivers State, Nigeria) and brine at various concentrations. The solutions were mixed using a magnetic stirrer until a homogeneous condition is attained, and then aged for 24 hours at room temperature (~30°C). After 24 hours, alumina nanoparticles (provided by department of petroleum and gas Engineering laboratory, university of Port Harcourt, Rivers State, Nigeria) were introduced into the solution and mixed until fully dissolved, resulting in a homogeneous solution. The pH values of each flooding fluid were also determined at different concentrations using a pH meter. Rheological parameters, including viscosity, were measured using a U-tube viscometer at the same temperature. The mutual correlation between different parameters were established to examine the impact of factors such as polymer and hybrid concentration on solution viscosity.

2.2.2 Sand Pack Flooding Experiment

The experimental procedure is as follows: The flooding experiment commenced with a drainage test; wherein crude oil was injected using the experimental set-up for flooding as shown in Figure 1. This process displaced the laboratory brine in the sand-pack until irreducible water saturation was achieved, indicated by the absence of water droplets through the outlet pipe into the measuring cylinder. The time at which the first drop of oil was observed through the outlet pipe, known as the oil breakthrough time, was recorded using a stopwatch. Additionally, the volume of brine displaced by the oil within the plug was estimated as the initial volume of oil in place (OIIP). Next, the imbibition test was conducted by injecting brine through the inlet pipe into the core holder to displace the oil until irreducible oil saturation was achieved. The volume of oil displaced after water flooding was recorded, and the time for water breakthrough was also noted. The efficiency of oil displacement was calculated and recorded. To prepare PAM and hybrid solutions, 100ml of brine (NaCl + KCl) with polymer (PAM) concentrations of 0.1wt%, 0.2wt%, and 0.3wt% respectively, along with alumina nanoparticles (Al_2O_3 NP), were used. The tertiary oil through the inlet pipe into the core-holder to displace additional oil, as depicted in Figure 1. The amount of oil recovered was recorded, and the efficiency of oil recovery was calculated and recorded as well. Subsequently, the unconsolidated sand pack was removed from the core holder and weighed. It is important to note that this process was repeated for each sand pack. Finally, the viscosity of each PAM solution and PAM/Al_2O_3 hybrid was evaluated. Table 1 presents the properties of the crude oil sample utilized in the flooding experiment.

Table 1: Properties of Crude Oil

Properties	Values
Specific gravity	0.860
Density (g/cc)	0.8880
Temperature (℃)	30
API gravity (°)	33.99
Viscosity (cP)	42.6812

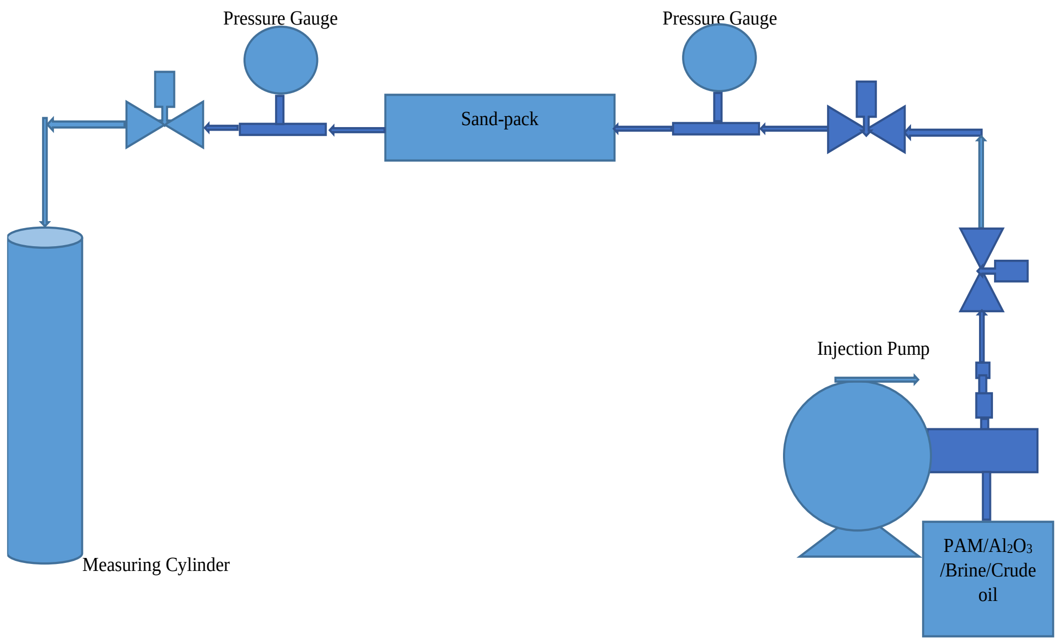

Figure 1: Experimental Apparatus design for the Flooding Process

3. Results and Discussion

3.1 Effect of PAM and Hybrid Concentration on the Viscosity of Solutions

This evaluation was carried out using a reference solution of 0.1 to 0.3 wt% PAM without alumina nanoparticles. The same concentrations of the PAM/Al_2O_3 hybrid dispersed in brine were also conducted for comparison. As the concentration of PAM in the solution increases, the viscosity of the solution also increases as shown in Figure 2. This is because, the polymer chains in PAM solutions becomes more entangled at higher concentration leading to a higher resistance to flow. Hence, increase in viscosity at increased polymer concentration confirms beneficial effect for a polymer flooding process since higher viscosity of the PAM solution results in increased oil recovery efficiency as shown in Figure 3 and Figure 4 respectively. An increase in viscosity of the polymeric solution due to increased polymer concentration was observed by Yadav et al. (2020), when they conducted a study on the experimental evaluation of partially hydrolyzed polyacrylamide and silica nanoparticles solutions for enhanced oil recovery [2].

The viscosity of the PAM/ Al_2O_3 hybrid solution was measured with varying hybrid concentration ranging from 0.1wt% to 0.3 wt% at constant brine concentration of 100ml and temperature of 30°C, to determine their effectiveness in increasing the mobility of the oil phase and improving fluid flow through the porous media. The results showed that from 0.1wt% to 0.3wt% concentration the PAM/ Al_2O_3 hybrid solution exhibited higher viscosity of 32.8964cP, 47.9557cP, and 57.19cP respectively compared to 25.9003cP, 41.5084cP, and 49.6815cP respectively obtained by the bare PAM solution. As the concentration of PAM/ Al_2O_3 hybrid increases, the viscosity increases, and surpasses that of PAM alone. This is as a result of the formation of hydrogen bonding between the aluminol groups in the alumina nanoparticles (Al_2O_3 NPs) and the amide groups in the PAM. The Al_2O_3 nanoparticles have high surface area and energy, leading to a strong interaction with the PAM molecules. This results in a more rigid and structured polymer network, which increases the viscosity of the solution. Furthermore, the Al_2O_3 nanoparticles act as physical barriers to the flow of polymer chains, thereby further increasing the viscosity of the solution. Hence, the addition of Al_2O_3 NPs to PAM solution creates a hybrid material with better mobility control, enhanced solution stability and improved rheological properties such as increased viscosity, which makes it more effective and suitable for enhanced oil recovery.

The increased viscosity of the PAM/Al_2O_3 hybrid solution helps improve displacement efficiency and reduce residual oil, resulting in enhanced oil recovery. The higher increase in cumulative oil recovery was observed as the viscosity increases as shown in Figure 3. It is evident from Figure 2 that the viscosity of the hybrid solution increases with higher alumina nanoparticle concentrations. As a result, it effectively displaces the oil during the polymer flooding process, especially at higher PAM/Al_2O_3 hybrid concentrations. This concept has been observed and investigated in previous literature by Ali et al. (2019) during the application of low salinity polymeric-nanofluids in carbonate oil reservoirs. The polymeric-nanofluid resulted in a higher oil recovery due to higher viscosity, significant reduction in interfacial tension, better emulsion stability, and wettability alteration towards a stronger water-wet system [16]. Table 2 illustrates that an increase in the concentration of the polyacrylamide (PAM) leads to a corresponding increase in the viscosity of the solution.

Table 2: Experimental Result of the Viscosity of the Fluid Samples

Dispersing Fluids	Temperature (℃)	Efflux time (Sec)	Density of the fluids (g/cc)	Viscometer constant 150/601B	Kinematic viscosity (m^2/Sec)	Dynamic viscosity(cP)
PAM1	30	701	1.0147	0.03641240	25.525092	25.9003
Hybrid1	30	890	1.0151	0.03641240	32.407036	32.8964
PAM2	30	1122	1.0160	0.03641240	40.854713	41.5084
Hybrid2	30	1295	1.0170	0.03641240	47.154058	47.9557
PAM3	30	1342	1.0167	0.03641240	48.865441	49.6815
Hybrid3	30	1543	1.0179	0.03641240	56.184333	57.1900
Brine (30,000ppm)	30	91	1.0174	0.03641240	3.3135284	3.3712
Crude oil	30	1320	0.8880	0.03641240	48.064368	42.6812

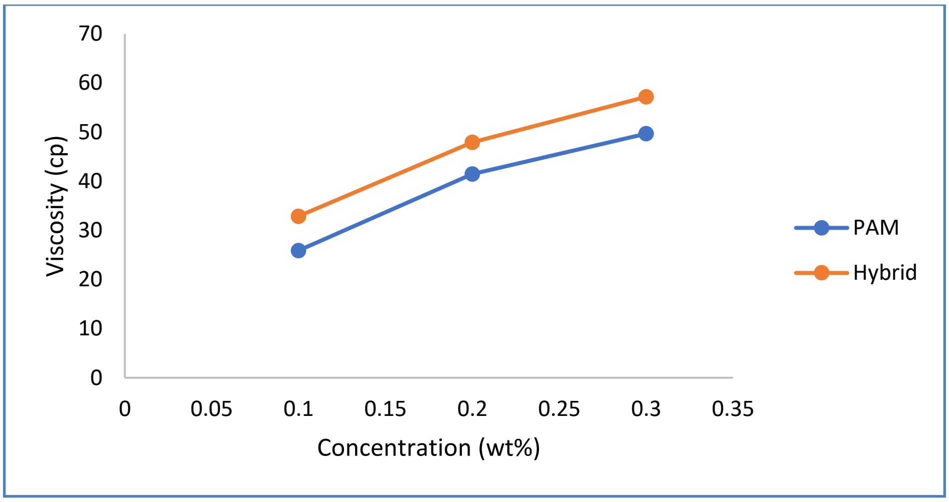

Figure 2: Correlation between PAM and Hybrid Concentration on the Viscosity of Solutions

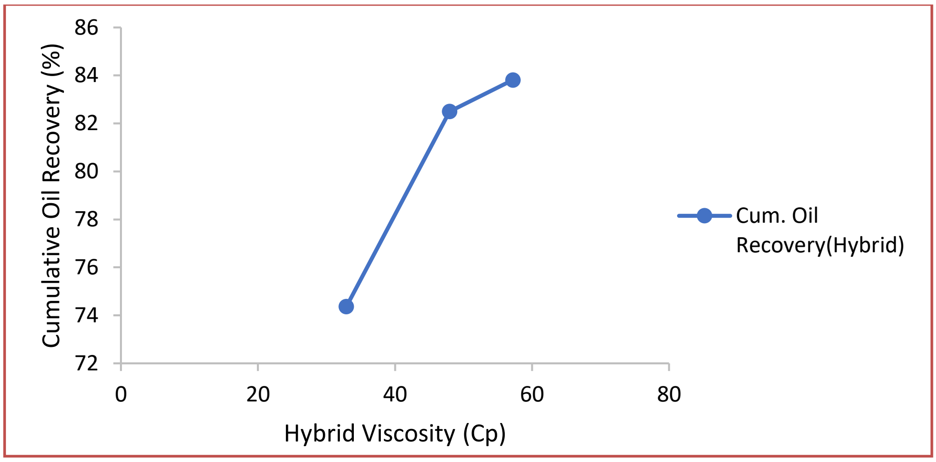

Figure 3: Cumulative Oil Recovery (%) as a function of Hybrid Viscosity

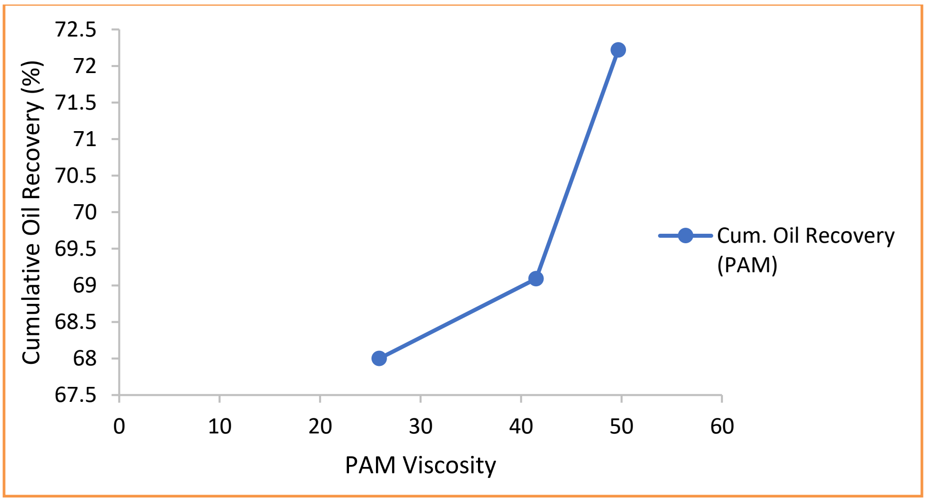

Figure 4: Cumulative Oil Recovery (%) as a Function of the Polymer Viscosity

3.2 Effect of PAM and Hybrid Concentration on the pH of the Solution

The pH of the solutions was determined by varying the concentration of PAM and PAM/Al_2O_3NP hybrid, ranging from 0.1wt% to 0.3wt%, at a brine concentration of 100ml. The results indicate that as the concentration of the solutions increases from 0.1 wt% to 0.3 wt%, the pH also increases. The hybrid solution demonstrates higher pH values of 7.1, 7.4, and 7.9 respectively, while the PAM solution exhibits lower pH values of 5.8, 6.0, and 6.1 respectively, as depicted in Figure 5. An increase in the cumulative oil recovery was also observed as the pH of the solution increased, with the hybrid solution resulting in higher recovery rates, as shown in Figure 6 and Figure 7. This is attributed to the enhanced basic nature of the PAM/Al_2O_3 hybrid due to the interaction between PAM and Al_2O_3 NPs, resulting in a greater increase in pH compared to PAM alone, thereby leading to a higher recovery rate. The presence of hydroxyl groups (OH-) on the surface of Al_2O_3 accepts protons to form water and hydroxide ions, thereby increasing the pH. The addition of Al_2O_3 nanoparticles to the PAM solution enhances its adsorption onto the rock surface and improves its wettability, consequently reducing interfacial tension and mobilizing trapped oil. Table 3 also demonstrates an increase in the pH value for both the PAM solution and PAM/ Al_2O_3 hybrid as the concentration increases.

Table 3: pH Values for Dispersing Fluid Samples

S/N	Dispersing Fluids	pH values
1	PAM1	5.8
2	Hybrid1	7.1
3	PAM2	6.0
4	Hybrid2	7.4
5	PAM3	6.1
6	Hybrid3	7.9
7	Brine (30,000ppm)	6.5

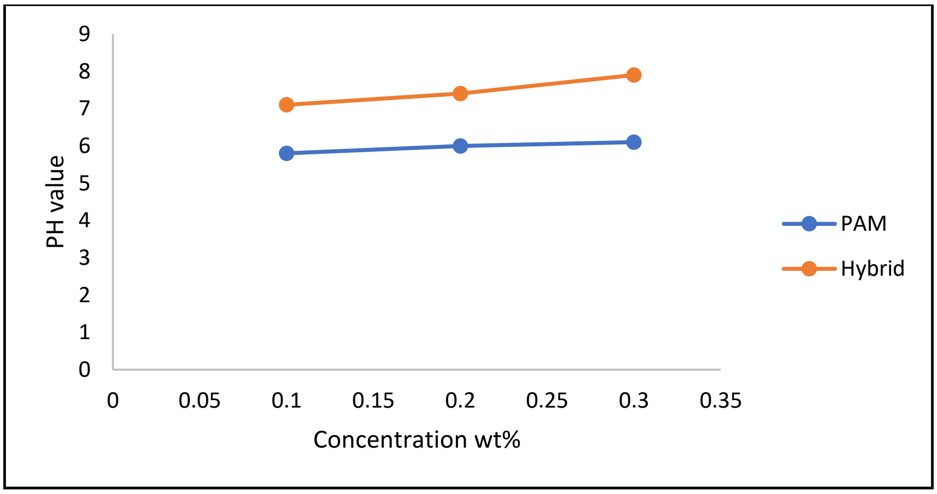

Figure 5: pH values versus PAM and Hybrid Concentration

Furthermore, the Al_2O_3 nanoparticles enhance the viscosity of the PAM solution, thereby improving sweep efficiency and reducing the amount of residual oil left in the reservoir. Additionally, the chemical properties of Al_2O_3 nanoparticles improve the stability of the PAM/Al_2O_3 hybrid solution, making it more effective in reducing interfacial tension and mobilizing the oil. Therefore, the addition of alumina nanoparticles increases the pH value of the PAM/Al_2O_3 NP hybrid, thereby enhancing the efficiency of oil removal compared to the bare PAM solution, as depicted in Figure 6 and Figure 7, respectively. This finding aligns with the study conducted by Rueda et al. (2020) on the experimental investigation of the effect of adding nanoparticles to polymer flooding in a water-wet micromodel. The study demonstrates that the addition of nanoparticles increased the pH of the hybrid solution by an average of 6.2% [17].

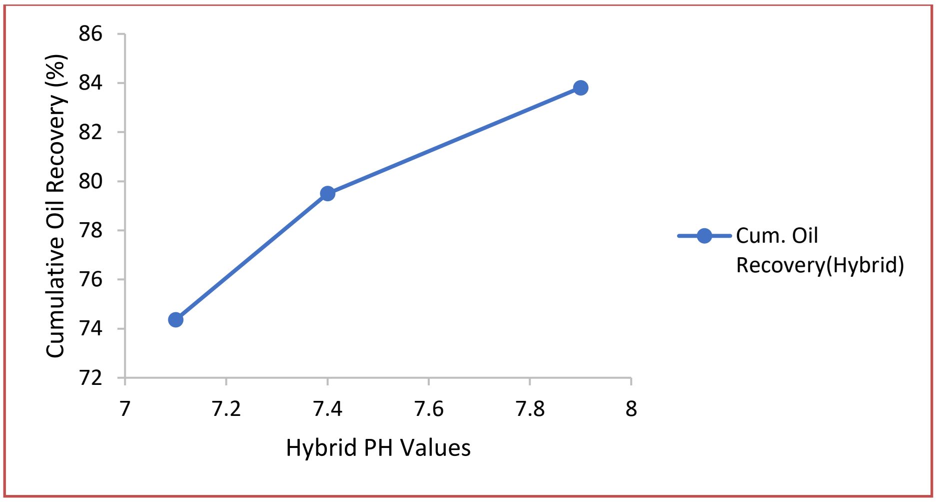

Figure 6: Effect of Hybrid pH on Cumulative Oil Recovery

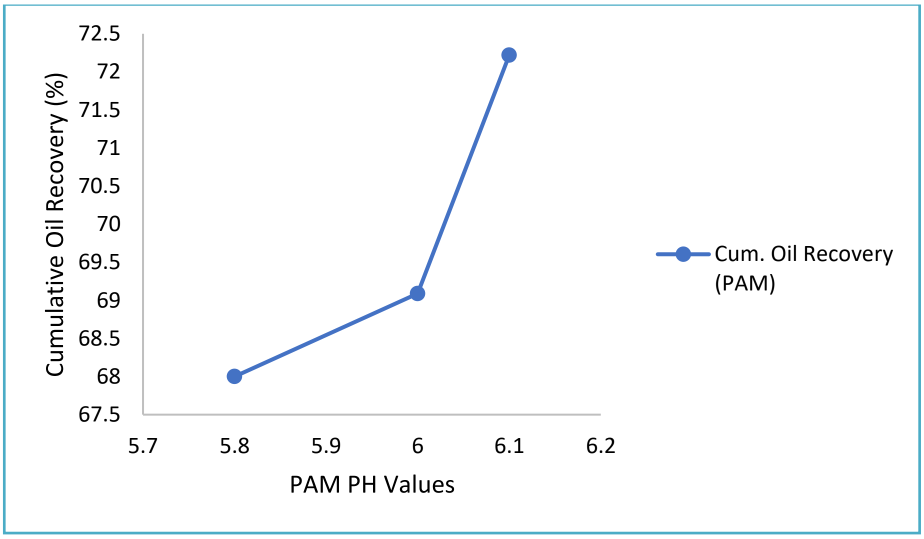

Figure 7: Effect of PAM pH on Cumulative Oil Recovery

3.3 Effect of PAM and Hybrid Concentration on Cumulative Oil Recovery

Chemical-enhanced oil recovery (CEOR) was conducted using a polyacrylamide (PAM) solution and a PAM/Al_2O_3 hybrid in a sand pack at a temperature of 30°C. The results of the hybrid flooding experiment were compared to those of the PAM solution alone after a waterflooding process. Subsequently, the PAM solution and PAM/Al_2O_3 hybrid were separately examined as tertiary recovery processes for oil recovery.

The PAM solution alone showed a slight increase in oil recovery after the waterflooding process, with a cumulative oil recovery ranging from 70.68% to 72.22% of the original oil in place (OOIP). In contrast, the PAM/Al_2O_3 hybrid demonstrated a better cumulative oil recovery, ranging from 74.36% to 83.81% OOIP, as the concentration increased from 0.1wt% to 0.3wt%. The increase in cumulative oil recovery was directly proportional to the concentration of the solution, as depicted in Figure 8. Table 4 illustrates that the PAM/Al_2O_3 hybrid outperformed the bare PAM solution in terms of oil recovery, particularly as the concentration increased. This can be attributed to the enhanced properties of the PAM/Al_2O_3 hybrid, such as viscosity, stability, and mobility control, which enable more efficient mobilization of the oil, particularly at higher fluid concentrations.

These findings align with a flooding experiment conducted by Yadav et al. (2019), which demonstrated that the addition of nanoparticles improves production rates [2]. The use of alumina nanoparticles (Al_2O_3 NPs) as an additive in this study enhanced the rheological properties of the PAM solution, controlled thermal degradation, and reduced chemical degradation due to their shielding effects. Moreover, the inherent surface-active nature of the alumina nanoparticles in the PAM/Al_2O_3 hybrid contributed to its superior efficiency. The hybrid exhibited enhanced thermal stability, higher rheological properties, and the potential to reduce oil viscosity, thereby enabling better conformance control and improved sweep efficiency.

Table 4: Summary of Cumulative Oil Recovery at 0.1-0.3wt% Concentration

Plug Sample ID	Dispersing Fluids	OIIP (ml)	Secondary Recovery (ml)	Tertiary Recovery(ml)	Cumulative Oil Recovery (ml)	Cumulative Oil Recovery (%)
A	PAM1	19.00	12	0.92	12.92	68.00
B	Hybrid1	19.50	12	2.50	14.50	74.36
C	PAM2	22.00	13	2.20	15.20	69.09
D	Hybrid2	20.00	12	4.50	16.50	82.50
E	PAM3	18.00	11	2.00	13.00	72.22
F	Hybrid3	21.00	12	5.60	17.60	83.81

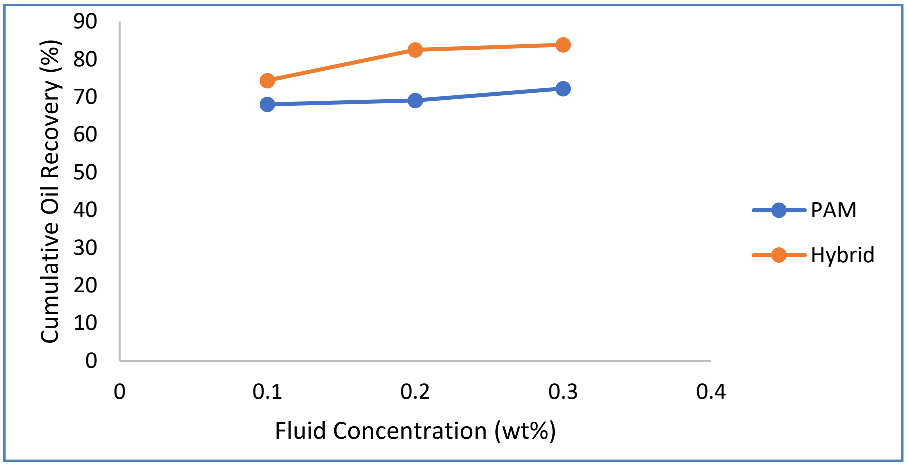

Figure 8: Cumulative oil recovery as a function of PAM and Hybrid concentration

3.4 Effect of the Efflux time on Cumulative Oil Recovery

Efflux time measurements were conducted to assess the flow dynamics of the solutions. The PAM/Al_2O_3 hybrid solution exhibited a longer efflux time, indicating its ability to maintain viscosity and provide sustained flow control. The increase in efflux time from 701 seconds to 1342 seconds resulted in a slight rise in cumulative oil recovery from 70.68% to 72.22% for the PAM solution alone. In contrast, a higher increase in cumulative oil recovery from 74.36% to 83.81% was observed for the PAM/Al_2O_3 hybrid solution with an increased efflux time from 890 seconds to 1543 seconds.

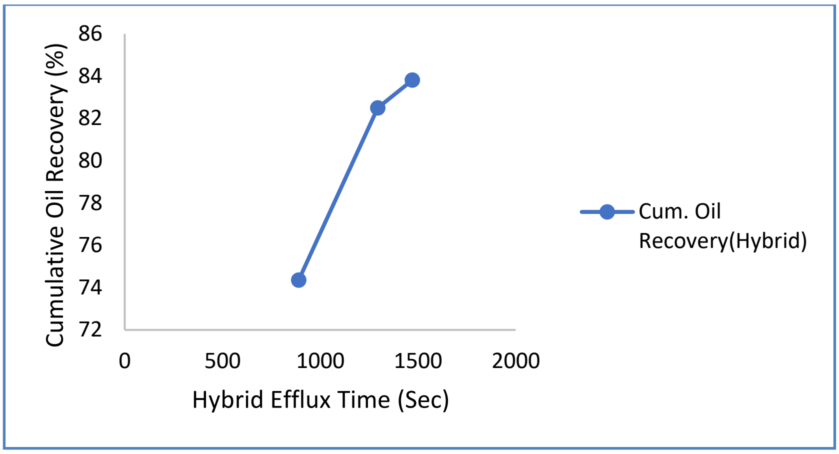

Figure 9: Cumulative Oil Recovery as a function of Hybrid Efflux Time

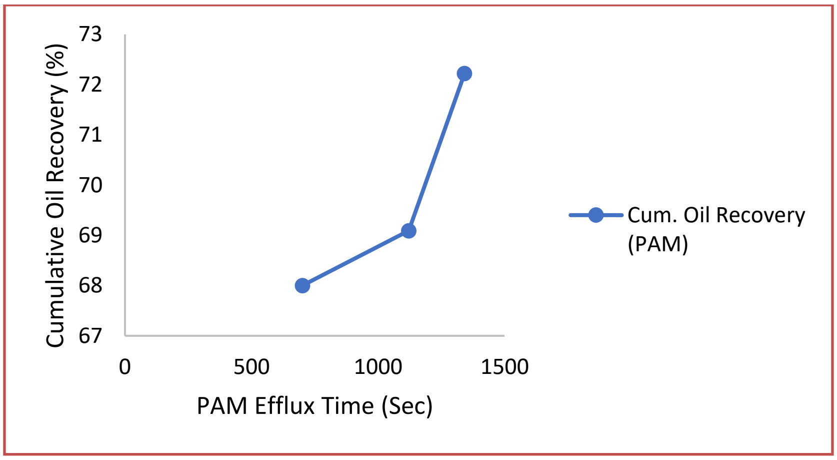

Figure 10: Cumulative Oil Recovery as a function of PAM Efflux Time

Figures 9 and Figure 10 demonstrate that the PAM/Al_2O_3 hybrid solution yielded superior cumulative oil recovery compared to the PAM solution as the efflux time increased. The enhanced cumulative oil recovery during PAM/Al_2O_3 hybrid flooding, as depicted in Figure 9, can be attributed to improved sweep efficiency and reduced mobility ratio resulting from the increased viscosity of the PAM/Al_2O_3 hybrid solution and the utilization of Al_2O_3 nanoparticles. This finding aligns with a study conducted by Odo et al. in 2020, which demonstrated that increasing the efflux time of Al_2O_3 nanoparticles leads to an increase in viscosity and subsequently improves oil recovery [1].

4. Conclusions

This study aimed to investigate and determine the suitability of alumina nanoparticles (Al_2O_3 NPs) as an additive to enhance the rheological and oil recovery properties of polyacrylamide (PAM) solutions. The effectiveness of PAM solutions and PAM/ Al_2O_3 hybrid solutions was evaluated and compared. The results demonstrated that the hybrid used in the chemical enhanced oil recovery (CEOR) experiment exhibited superior rheological properties compared to the pure PAM solution. Specifically, the hybrid solution had a viscosity of 57.19 cP at a concentration of 0.3 wt%, while the pure PAM solution had a viscosity of 49.68 cP. Additionally, flooding with the PAM/Al_2O_3 hybrid resulted in higher oil recovery efficiency compared to flooding with the pure PAM solution, particularly as the concentration increased. This improvement can be attributed to the addition of Al_2O_3 to the PAM solution, which is used to displace the oil.To evaluate oil recovery, three concentrations of PAM polymer and PAM/Al_2O_3 hybrid ranging from 0.1 to 0.3 wt% were utilized. It was observed that the PAM/Al_2O_3 hybrid achieved the best result at a concentration of 0.3 wt%, with an oil recovery efficiency of 83.81%. In comparison, the pure PAM solution achieved an oil recovery efficiency of 72.22% at the same concentration. To compare the efficiency of dispersing fluids such as PAM and hybrid solutions, a statistical model was used to generate the mutual correlation between viscosity, pH, efflux time, concentration, and oil recovery efficiency of the pure PAM and hybrid solutions.

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