

**INVESTIGATION OF SILICA NANOPARTICLE-POLYMER HYBRID  
STABILITY UNDER HIGH TEMPERATURE AND SALINITY FOR OIL  
DISPLACEMENT APPLICATION**

**AMER ISKANDAR BIN RA'EIS**

**UNIVERSITI TEKNOLOGI MALAYSIA**

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DISPLACEMENT APPLICATION

AMER ISKANDAR BIN RA'EIS

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## **DEDICATION**

This thesis is dedicated to Allah s.w.t, my creator, tower of strength, and source of inspiration, wisdom, knowledge, and comprehension. He has been my source of strength throughout this program. This work is also dedicated to my parents, Ra'eis Aziz and Norliza Jamil, who have always loved me unconditionally and whose excellent example led me to work hard for my goals. To my buddy Faiz Rahim, Nurazam Talib and Zaki Hafizi, who are my source of inspiration and support during this Master programme. Sincere gratitude for your presence in my life. To my siblings Ammar and Aina, who have been influenced in every conceivable manner by our adventure. Thank you. My affection for you all is incalculable. God bless you.

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## ABSTRACT

Polymer flooding is one of the most often utilised enhanced oil recovery (EOR) techniques because it provides excellent recovery. Polymer flooding enhances sweep efficiency and reduce viscous fingering severity by increasing fluid and oil mobility. Due to excellent viscosifying nature, and well-known physiochemical properties, partially hydrolyzed polyacrylamide (HPAM) is the polymer most often utilised for the application. However, high temperatures restrict its application because polymer will acts as shear-thinning, such it undergoes shear degradation and reduces viscosity at high shear rates and quickly destabilized and therefore unable to achieve the expected effects. High salinity also causes the molecular chain of the polymer to collapse, which results in a much smaller molecule and hence, produces a lower viscosity solution. Adding nanoparticle to polymer solutions is now required to alter their properties. Therefore, this study aims to investigate the effect of silicon dioxide nanoparticles ( $\text{SiO}_2$ ) addition to the stability of HPAM at high temperatures and salinity. The shear viscosity and the flooding performance at high temperature and high salinity gauge the stability of HPAM and the hybrid HPAM-  $\text{SiO}_2$ . A series of stability measurements as well as core flooding experiment with variations of conditions were conducted in order to know the improvement offered by this nanoparticle towards HPAM polymer. At a temperature of 110 °C, the addition of 1 wt%  $\text{SiO}_2$  nanoparticle have enhanced the viscosity of 0.015 wt% HPAM, from 3.4 cP to 6.8 cP. This resulted in an almost 90% oil recovery rate. It also strengthened HPAM's salt tolerance at concentration of 5 wt% of NaCl by raising its viscosity up to 4.6 cP. This HPAM hybrid also have improve the oil recovery factor for this condition as well up to 85%. In conclusion, adding nanoparticles to HPAM will unquestionably increase the stability and potentially be used in EOR operations.

## ABSTRAK

Banjir polimer adalah salah satu teknik pemulihan minyak yang dipertingkatkan dan ianya paling kerap digunakan kerana ia memberikan kadar pemulihan yang sangat baik. Banjir polimer dapat meningkatkan kecekapan sapuan dan mengurangkan keterukan penjarian likat dengan meningkatkan mobiliti cecair dan minyak. Disebabkan sifat kelikatan yang sangat baik, dan sifat fisiokimia yang terkenal, *partially hydrolyzed polyacrylamide* (HPAM) ialah polimer yang paling kerap digunakan untuk setiap aplikasi. Walau bagaimanapun, suhu tinggi menyekat penggunaannya kerana polimer akan menipis, sehingga ia mengalami degradasi dan mengurangkan kelikatan pada kadar ricih yang tinggi dan tidak akan stabil dengan cepat. Oleh itu, ianya tidak dapat mencapai kesan yang diharapkan. Kemasinan yang tinggi juga menyebabkan rantai molekul polimer akan runtuh, justeru menghasilkan molekul yang lebih kecil dan larutan kelikatan yang lebih rendah. Penambahan nanopartikel kepada larutan polimer kini diperlukan untuk mengubah sifatnya. Oleh itu, kajian ini bertujuan untuk menyiasat kesan penambahan nanozarah seperti silikon dioksida ( $\text{SiO}_2$ ) terhadap kestabilan HPAM pada suhu dan kemasinan yang tinggi. Kelikatan dan prestasi banjir pada suhu tinggi dan kemasinan tinggi akan mempengaruhi kestabilan HPAM dan hibrid HPAM-  $\text{SiO}_2$ . Satu siri pengukuran kestabilan serta eksperimen banjir teras dengan variasi keadaan telah dijalankan untuk mengetahui peningkatan yang ditawarkan oleh nanopartikel ini terhadap polimer HPAM. Hasil menunjukkan bahawa, pada suhu 110 °C, penambahan 1 wt%  $\text{SiO}_2$  nanopartikel telah meningkatkan kelikatan 0.015 wt% HPAM daripada 3.4 cP kepada 6.8 cP. Ianya juga menyebabkan kadar pemulihan minyak meningkat sehingga hampir 90%. Penambahan nanopartikel ini juga telah mengukuhkan toleransi HPAM terhadap kemasinan, yakni pada kepekatan 5 wt% NaCl dengan peningkatan kelikatan sehingga 4.6 cP, dan seterusnya pengabungan HPAM hibrid ini juga telah meningkatkan faktor pemulihan minyak untuk situasi ini sehingga hampir 85%. Kesimpulannya, penambahan nanopartikel pada HPAM sudah pasti akan meningkatkan kestabilan dan berpotensi digunakan dalam operasi EOR.

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## LIST OF ABBREVIATIONS

<b>EOR</b>	–	Enhanced Oil Recovery
<b>HPAM</b>	–	Hydrolysed Polyacrylamide
<b>WC</b>	–	Water Cut
<b>IPV</b>	–	Inaccessible Pore Volume
<b>WAG</b>	–	Water Alternate Gas
<b>OOIP</b>	–	Original Oil In Place
<b>IGIP</b>	–	Initial Gas In Place
<b>OGIP</b>	–	Original Gas In Place
<b>VAPEX</b>	–	Vapour Extraction
<b>SAGD</b>	–	Steam Assisted Gravity Drainage
<b>AS</b>	–	Alkaline Surfactant
<b>SP</b>	–	Surfactant Polymer
<b>AP</b>	–	Alkaline Polymer
<b>ASP</b>	–	Alkaline Surfactant Polymer
<b>C</b>	–	Chromophore
<b>CCP</b>	–	Cationic-Conjugated Polymer
<b>RF</b>	–	Recovery Factor
<b>EUR</b>	–	Estimated Ultimate Recovery
<b>TOC</b>	–	Total Organic Content
<b>NP</b>	–	Nanoparticles

## LIST OF SYMBOLS

$F_R$	-	resistance factor [f]
$F_{RR}$	-	residual resistance factor [f]
$\lambda_w$	-	brine mobility, [ $\mu\text{m}^2/(\text{mPa}\cdot\text{s})$ ]
$\lambda_p$	-	polymer solution mobility, [ $\mu\text{m}^2/(\text{mPa}\cdot\text{s})$ ]
$k_w$	-	effective permeability of brine, [ $\mu\text{m}^2$ ]
$k_p$	-	effective permeability of polymer solution [ $\mu\text{m}^2$ ]
$u_w$	-	viscosity of brine, [mPa.s]
$u_p$	-	viscosity of polymer solution, [mPa.s]
$Q$	-	flow rate through the core, [ $\text{cm}^3/\text{s}$ ]
$L$	-	length of the core, [cm]
$A$	-	section area of the core, [ $\text{cm}^2$ ]
$\Delta p$	-	pressure difference between two ends of the core, [atm]
$k_{wi}$	-	brine permeability before polymer flows through the core, [ $\mu\text{m}^2$ ]
$k_{wa}$	-	brine permeability after polymer flows through the core, [ $\mu\text{m}^2$ ]
$k$	-	pore permeability, [ $\mu\text{m}^2$ ]

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# CHAPTER 1

## INTRODUCTION

### 1.1 Background of Research

There are three stages of oil recovery. The first stage will utilize the pressure within the reservoir to extract the oil. Inevitably, the reservoir's pressure will eventually decline, resulting in unsustainable oil production. Secondary stages include gas injection, water flooding and water injection, with the primary objective being of maintaining the reservoir pressure. Finally, tertiary or enhanced oil recovery (EOR) is always employed to improve the flooding process area and volumetric sweep efficiency. The three most commonly implemented EOR includes thermal EOR, chemical EOR, and gas miscible EOR. EOR overcomes well-known causes of inefficient flooding processes such as viscous fingering, bypassed oil, segregated flow, and altered reservoir wettability (Ali Mohsenatabar Firozjaiia and Hamid Reza Saghafi, 2020).

Chemically enhanced oil recovery improves volumetric sweep efficiency by lowering the water-oil mobility ratio such as polymer and decreases residual oil saturation by reducing the water-oil interfacial tension including surfactant or alkaline. Chemical flooding EOR comes in various forms, including alkaline flooding, surfactant flooding, surfactant-polymer flooding, polymer flooding, and alkaline-surfactant-polymer flooding. Theoretically, a water-soluble polymer raises the water viscosity and reduces the water-oil mobility ratio, improving the volumetric sweep's effectiveness (Ali Mohsenatabar Firozjaiia, 2020).

One of the most often used EOR methods is polymer flooding because it offers excellent recovery. Its objective is to improve sweep efficiency and lessen the severity of viscous fingering by decreasing the mobility ratio between the displacing fluid and the oil. The ability of the polymer to increase viscosity at a low concentration is the main emphasis of the design. The oil recovery improves as much as 15 to 20 percent for polymer compared to conventional water flooding. Polymer flooding works exceptionally in highly heterogeneous reservoirs where the viscous fingering is severe and in high-viscosity resident oil reservoirs where the mobility ratio is more than unity (Hamid Reza Saghafi, 2020).

Viscous fingering is the unstable displacement of viscous fluid by a less viscous fluid. Polymer flooding is frequently employed to combat viscous fingering. The fingering of injection fluid may harm the reservoir's flow, and the oil recovery of a reservoir. One reasons for viscous fingering is the reservoir rock's heterogeneous permeability. Water or another displacing fluid will only flow through the highly permeable layer, leaving the lower permeability layer's oil unaffected. Fluid viscosity, rock heterogeneity, temperature, and varying injection rates all contribute to viscous fingering. Gravity segregation, which separates the denser water from the oil, is a frequent term for this process. Combination of gravity segregation and viscous fingering results in the early water breakthrough and poor area and volumetric sweep efficiency result (Sameer Al-Hajria and Maziyar Sabet, 2019).

A detailed investigation of the polymer solution has been conducted to handle the significant flooding problem. A polymer solution practically increase the water's viscosity, leading to a stronger viscous force and higher sweep efficiency. Large molecules known as polymers are made up of several monomers, or smaller chemical building blocks (Karl D, 2021). The two kinds of polymers are biopolymers and synthetic polymers. Synthetic polymers are huge molecular chains (macromolecules) and organic connections produced from natural products or by synthesising basic

materials from oil, gas, or coal, such as hydrolysed polyacrylamide (HPAM). Biopolymers are organic polymers created by the cells of living things, such as chitosan. In biopolymers, monomeric units are covalently bonded to produce further enormous molecules. The three main categories of biopolymers are polynucleotides, polypeptides, and polysaccharides and these categories are based on the monomers used and the structure of the biopolymer produced (Md Irfan, and Christopher P. Lenn, 2021)

According to E. Mentzer, J. Heemskerk, and E.J.L. Koning (2017), the Marmul field in south Oman's sandstone reservoirs benefits from HPAM flooding, which lowers the total water-cut from 50% to 20%. Reducing water-cut extend the life of wells and save maintenance costs for surface infrastructure like separators. N. Sanmartim (2002) reported that the oil viscosity of 50 cP at 50°C after studying the polymer flooding process using HPAM at the Canto do Amaro oil field in Brazil. This outcome demonstrates the viability of polymer flooding in a reservoir containing high-viscosity oil. However, the use of polymer flooding is restricted by high salinity and high temperatures reservoirs, leading to several studies to enhance the HPAM's stability.

## **1.2 Problem Statement**

Due to its cheap cost, tendency to viscosify, and well-known physiochemical features, partially hydrolysed polyacrylamide (HPAM) seems to be the most commonly used polymer in this situation. However, reservoir's high temperature and salinity restrict the use of HPAM. The chain's amide groups undergo substantial hydrolysis into a carboxylic acid in reservoir conditions. When these hydrolyzed products come in touch with the common cations found in reservoir brines, they precipitate. HPAM behaves as a shear-thinning polymer, reducing viscosity and going through shear degradation at high shear rates. This is because the polymer chains are

severed at high shear rates (Song, 2006). Dandan Hu (2014) stated that mostly the limits for polymer flooding are that viscosity is lower than 100cP, the temperature is lower than 93 °C, water salinity is lower than 100,000 ppm, and formation permeability is higher than 20mD. In this high temperature and salinity environment, this polymer is quickly destabilized and unable to achieve the expected effects. High salinity also causes the polymer's molecular chain to collapse, producing a much smaller molecule and a lower viscosity solution.

Thermal and salinity degradation both impact the rheology behaviour of polymer solutions in porous media. The rate at which the polymer solution is hydrolyzed is directly influenced by temperature. Polymer flooding often resulted in HPAM hydrolyzing between 25 and 90 degrees. The most often utilised compounds in oilfield applications are partly hydrolyzed polyacrylamide (HPAM) and water-soluble polyacrylamide (PAM). HPAM polymers are often employed as mobility-controlling agents to raise the viscosity of stimulation fluids in polymer-augmented water flooding enhanced oil recovery (EOR) procedures to boost sweep efficiency. However, due to the thermal instability, applications of HPAM more than 90 °C and high salinity in between 100,000 and 200,000 ppm reservoir conditions are severely constrained. According to Algharaib M. (2014), this hydrolysis phenomenon may also decrease in the viscosity of a polymer solution. The viscosity of a polymer solution is influenced by several variables, including the concentration of the polymer, the rate at which it degrades, the temperature, as well as the salinity of the water used to prepare the solution.

The concentration of a dissolved polymer also affects the viscosity of the solution. The polymer segments tend to be surrounded with solvent molecules in appropriate solvents over other polymer segments. The polymer molecules, on the other hand, work to reduce the region of contact well with solvent molecules in poor solvents. Al-Zahrani (1990) explains that the extension of the polymer molecule, for instance, relies on the interaction of a polymer solvent, which has an immediate impact on its size as well as the viscosity of the solution.

Nowadays, adding nanoparticles into polymer solutions might be of interest, but more research on the viability of nanoparticles including the effects on oil recovery is still needed. The enhancement that  $\text{SiO}_2$  offers to HPAM stability under high temperatures and high salinities, as well as the impact of these polymer hybrids on HPAM flooding performance, are nonetheless things that should be investigated.

### **1.3 Research objectives**

The primary objective is to investigate the effect of silica nanoparticles addition on HPAM stability under high salinity and temperature.

1. To investigate the improvement silica dioxide nanoparticles ( $\text{SiO}_2$ ) to the HPAM stability in high temperature and high salinity.
2. To investigate the effect of silica dioxide nanoparticles ( $\text{SiO}_2$ ) on HPAM flooding performance under high temperature and high salinity.

## **1.4 Scope of Research**

### **1.4.1 Nanoparticle Preparation**

1. 100 ml of distilled water will be mixed with three different concentrations of HPAM such as 500, 1000, and 1500 ppm.
2. Stir all the HPAM solution at 500 rpm for 2 hours using a magnetic stirrer for homogeneous solution.
3. Measure the shear viscosity of all HPAM solution using Brookfield Rheometer at a shear rate of  $100s^{-1}$  for comparison matter.
4. The best viscosity result from the comparison will be selected, which justify as optimum performance for HPAM.
5. Prepare the selected HPAM concentration mixed with three different  $SiO_2$  solutions such as 0.01, 0.1, and 1wt% by repeating all the previous preparation method.
6. The best viscosity result from the comparison will be selected, which justify as optimum performance for HPAM-  $SiO_2$ .

### **1.4.2 Temperature and Salinity Experiment for HPAM Solution**

1. Heat the chosen HPAM solution at three different temperatures such as  $50^{\circ}C$ ,  $70^{\circ}C$ , and  $90^{\circ}C$  for 24 hours and measure the shear viscosity using increasing shear rate from 100 to  $1000s^{-1}$  to determine the effect of temperature to the HPAM solution.
2. Prepare HPAM solution with additional of 20,000, 50,000, and 80,000 ppm of sodium chloride (NaCl) and measure the shear viscosity using

increasing shear rate from 100 to  $1000s^{-1}$  to determine the effect of salinity to the HPAM solution stability.

3. All the process is repeated three times, and the average value is taken to ensure the consistent results.

#### **1.4.3 Temperature and Salinity comparison between HPAM with HPAM-SiO<sub>2</sub> solution**

1. Repeat all the experiments, which include temperature and salinity procedures with HPAM- SiO<sub>2</sub> solution
2. Compare the viscosity result on the worst temperature and salinity result made by HPAM solution previously for improvement matters.

#### **1.4.4 Flooding performance Experiment**

1. Prepare the several core samples using the glass beads as the core and the cylindrical column as a holder with 18 cm long and 3.8 cm inner diameter.
2. The porosity and permeability of each core were measured.
3. The core was saturated for 24 hours.
4. From the existing column made previously (sand + water), oil will be injected until it reaches the end of the column and some will come out with certain amount of water, then that amount of water will be recorded for finding irrisidual water saturation, Swirr.



5. For finding the effective temperature, the test material (Water, HPAM, HPAM + SiO<sub>2</sub>) will be heated up to the chosen temperature which is 110 degrees Celsius and it will be injected to the core model.
6. Same for effective salinity, test material (water, HPAM, HPAM + SiO<sub>2</sub>) will be mixed with the highest chosen concentration which is 50,000 ppm of NaCl and it will be injected to the core model.
7. Data for Pressure change ( $\Delta P$ ) and Volume of oil out (Vo out) will be recorded in order to plot the graph of Resistance and Recovery Factor.

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