Performance Evaluation of Viscosity Characteristics of Enhanced Preformed Particle Gels (PPGs)

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ABSTRACT: Preformed Particle Gels (PPGs) treatment is one of the most promising solutions to improve conformance control in mature water flooded oil reservoirs. It is very important to be able to evaluate and predict performance of PPGs in porous media. In this paper, we will first introduce new class of enhanced PPGs designed for harsh reservoir conditions (high salinity and temperature) and then present experimental laboratory results looking at viscosity performance of PPGs during injection and flow in porous media. A central composite experimental design coupled with a simple method using a set of 150-cm tubes, were used to comprehensively evaluate the functionality of PPGs rheological behavior under different flow conditions in fractures. The effects of five variables including salinity of water used to prepare the swollen PPGs, tube internal radius, injection velocity, size of PPGs and temperature on the PPGs viscosity were examined. The results showed that PPGs viscosity primarily depends on the injection velocity, tube internal radius, temperature, PPG size, salinity and their two-level interactions. It is also worth to mention that the effect of temperature on PPGs viscosity has not been investigated in previous studies. Finally, a simple mathematical model was introduced to predict the PPGs viscosity at reservoir conditions

KEYWORDS: Preformed particle gels; Conformance control; Viscosity; Experimental design.

INTRODUCTION

Due to mineral dissolution, oil reservoirs with induced fractures or high-permeability channels are quite common in mature oilfields [1, 2]. The wide variation in the permeability of these oil reservoirs causes uneven

water flooding during oil displacement process, resulting in poor reservoir sweep efficiency and very early high water breakthrough in producing wells. High water cut increases costs related to scale, corrosion, and water/oil

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separation, and makes a well unproductive and economically inefficient [3, 4]. Therefore, improving water flooding sweep efficiency has been the subject of many studies during the past years [5-8].

In-situ cross-linked polymer gel is traditionally one of the most commonly applied technologies for conformance control [9-14]; however, uncontrolled gelation times and variations in gelation time due to shear degradation as well as gelant compositional changes induced by contact with reservoir minerals and fluids are distinct drawbacks of this method [12, 13]. In addition, in-situ gelation systems behave as a polymer solution before gelation. During a polymer flooding process, polymer solution will also enter more in the zones un-swept by water during water flooding. Once gelant forms gel in un-swept zones, it will seriously damage the potential oil production of those zones [15].

A more recent approach that does not have the above mentioned disadvantages is Preformed Particle Gels (PPGs) which are expandable and hydrophilic polymeric particles. PPGs are a powder product made up of a cross-linked polymer that swells after addition to injection brine. The PPGs are added to the injection water for some period of time, and then followed by normal water injection. Advantages of this technology over traditional in-situ gel include: PPGs are environmentally-friendly, strength- and size-controlled materials, have adjustable mechanical properties and adjustable swelling ratios, and are stable in almost all formation water salinities. PPGs have only one component during the injection and can be prepared with produced water. Also, they can preferentially enter into fractures or fracture-feature channels while minimizing gel penetration into low permeable hydrocarbon zones/matrix [2, 16, 17]. Field applications of PPGs have had very positive results [2]; and PPGs have been applied in more than 4,000 wells in China to reduce fluid channels in both water flooding and polymer flooding operations [18].

Although most PPG-based treatments have been successfully applied in mature fields, there are only a few reports on PPGs viscosity evaluation. *Muhammed et al.* used a stainless steel screen plate with multiple holes to study the mechanism of passing swollen PPGs through the plate holes. They measured the stabilized extrusion pressure as a function of the flow rate and evaluated gel rheology in terms of its apparent viscosity as a function

of the shear rate [19]. Another study considered the shear-thinning properties of PPGs [20], in which theoretical mathematical models using the general power law equation, were developed to predict the pressure gradient of swollen PPGs during its extrusion through fractures. Then, these models were modified to predict the effective viscosity of swollen PPGs. The results show that the effective viscosity of PPGs decreases with an increase in injection rate and increases with an increase in fracture width. In another study of PPGs extrusion through opening conduits [7], some models were developed to quantitatively calculate the viscosity and the stable injection pressure as a function of the particle strength, particle-opening ratio and shear rate. They found that PPGs strength impacted injectivity more significantly than did particle-opening ratio, and the viscosity increased as the brine concentration and conduit opening size increased. These models correlated effective viscosity without considering the temperature effect.

The work presented here focuses on investigating the rheological behavior of these novel PPGs and to see how varying different parameters such as temperature, tube internal radius, salinity of water used to prepare the swollen PPGs, size of PPGs and injection velocity will affect the viscosity of PPGs. Finally a simple mathematical model is introduced to predict the PPGs viscosity at reservoir conditions.

EXPERIMENTAL SECTION

Materials

PPGs

The monomers used to prepare these enhanced PPGs were acrylamide (AM) (purity of 98.5%), N,N-dimethyl acrylamide (DA) (purity of 98.5%), 2-acrylamido-2-methylpropane sulfonic sodium salt (AMPSNa) (purity of 99%), N-vinylpyrrolidone (NVP) (purity of 98%), and the N,N'-methylenebis (acrylamide) (MBA) (purity of 99%) as a crosslinking agent. N,N',N,N' tetramethylethylenediamine (purity of 99%) and sodium persulfate were used as catalyst and initiator for polymer gel synthesis. All above mentioned chemicals were purchased from Beijing Chemical (Beijing, China). The nano clay montmorillonite Na+ used as a mechanical properties modifier was obtained from Aldrich (St Louis, MO, USA) and the nitrogen was obtained from Delvar Afzar Gas Industrial Group at 99.995% purity.

Table 1: Specifications of tubes used in viscosity test	Table 1:	Specification	ons of tubes	used in v	iscosity tests
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Tube Outside Diameter (mm)	Length (cm)	Internal Volume (cm ³)	Inside Radius (mm)	Thickness (mm)	Permeability (µm²)
1.59	150	0.48	0.32	0.48	398.2
3.18	150	1.27	0.52	1.07	1031.4
6.35	150	23.00	2.21	0.97	19085.2



Fig. 1: Dried PPGs

PPGs were prepared by free-radical polymerization of 2:1:1:2 molar mass ratio of AM, DA, NVP and AMPSNa monomers with 0.55 wt.% MBA and 2.5 wt.% nano clay montmorillonite Na+ at room temperature (300K) in distilled water. The manufacturing process was as follows: Nano clay montmorillonite Na+ added to the required amount of distilled water, followed by placing the mixer in an ultrasonic bath for 4 hours under continuous irradiation. Then specific amounts of AM and NVP monomers are weighed and sprinkled gently into the dispersed clay solution while stirring the solution. Then DA and AMPSNa monomer and a specific temperature stability agent were added. Stirring was continued for one hour until all the monomers were completely dispersed. Weighed MBA were poured into the solution while stirring vigorously and nitrogen purging were used. After stirring for 40 minutes, specific amounts of Sodium persulfate and the N,N,N',N'tetramethylethylenediamine catalyst were added and then nitrogen purging was stopped. The exothermic polymerization reaction began almost immediately, and hydrogel formation was observed within 30 minutes. PPGs were obtained by cutting the resulting gel into small pieces and drying it in an oven at 328K under vacuum condition for 24 hours (see Fig. 1).

Tubes

Three stainless steel tubes of various internal radiuses were selected for viscosity tests; their internal radii were 0.32, 0.52 and 2.21 mm. Each tube was cut into 150 cm long equal lengths. One side of each tube was equipped with a suitable connection. The permeability of each tube was calculated by Darcy law. Their specifications are shown in Table 1.

Experimental approach

Experimental set up

The set up consisted of a Vinci positive displacement pump (dual pump series), recombine transfer vessel for PPGs solution, brine transfer vessel, pressure transducer, data acquisition system and PC (see Fig. 2). The operational specifications of Vinci pump are shown in Table 2.

Tests methodology

The preparation of experimental materials and apparatus for viscosity tests were as follows:

- 1- The PPGs are placed in the water with the specified salinity at room temperature until they are completely swollen.
- 2- Excess brine solution should be separated from the swollen particle gels.
- 3- The swollen PPGs are packed in the transfer vessel which is equipped with a piston.
- 4- Two screw caps, equipped with an inlet and outlet nozzle, are used to pack the transfer vessel.
- 5- The top of the transfer vessel is connected to the discharge of the pump and the bottom is connected to the tube.
- 6- Transfer vessel and tube are placed in the oven at the test temperature for one hour before the test starts.
- 7- A pressure transducer is connected to the inlet of the tube to record the pressure data over time.

Once the apparatus is setup, the following experimental procedure is performed for each test:

Item	Unit	Range
Pressure	Pa	0, 100e+6
Flow	cm ³ /s	1.667e-5, 0.775
Operating temperature	К	278-473
Displacement resolution	mm ³	0.1

Table 2: Specifications of Vinci positive displacement pump.



Fig. 2: The schematic diagram of the experimental set up- Viscosity Test.

- 1- Initially, the pump is run at the maximum injection rate considered for that particular tube. The injection rates are designed such that we have equal injection velocities for all tubes.
- 2- As the PPGs are injected, the pressure buildup data is recorded every 5 seconds via the data acquisition system.
 - 3- Pressure data are recorded until pressure stabilized.
- 4- Once the pressure is stabilized the pump injection rate is lowered to the next lower injection rate and the pressure drop decline data are recorded.
- 5- This procedure is repeated for different injection rates.

To avoid error, no air gap should be left in downstream of the set up equipment.

The resistance factor is defined as the ratio of the PPGs injection pressure drop to the water injection

pressure drop at the same flow rate. The pressure during the PPGs injection was recorded and the pressure drop during water injection was calculated using the Poiseuille's law, which is as follows [15]:

$$\Delta P_{\text{brine}} = -257.312 \times 10^{-2} \times \left(\frac{\mu LV}{r^2}\right) \tag{1}$$

So, the resistance factor is defined as:

$$RF = \frac{\Delta P_{PPGs}}{\Delta P_{brine}} = \frac{\left(\frac{k}{\mu}\right)_{brine}}{\left(\frac{k}{\mu}\right)_{PPGs}}$$
(2)

Where $\left(\frac{k}{\mu}\right)_{\text{brine}}$ is the brine mobility during water

injection and $\left(\frac{k}{\mu}\right)_{\text{PPGs}}$ is the gel mobility during PPGs injection.

Parameters Unit Min. (-) Center or average (0) Max. (+) Gel Particle size μm 53.5 68.5 89.5 Water Salinity (NaCl) Molality 0 1.75 3.5 Tube inside radius mm 0.32 0.52 2.21 0.04 3.52 7.0 mm/s Injection Velocity 293.15 Temperature K 330.65 368.15

Table 3: Parameters constraints for Response surface design.

The permeability of the tube model remains the same before PPGs placement and during placement, so the resistance factor can be calculated as the ratio of PPGs effective viscosity divided by brine viscosity. Consequently, PPGs effective viscosity can be calculated as follows:

$$\mu_{eff,PPGs} = \mu_{eff,brine} \times RF \tag{3}$$

Where effective viscosity of brine depends on brine concentration, pressure, and temperature which can be found in the literature [21].

Design of Experiment

For viscosity tests, laboratory measurements were combined with an experimental design technique to screen effective parameters and develop quadratic response surfaces that can predict the viscosity of PPGs extrusion through tubes based on five controllable parameters and their interactions. The most popular response surface design is the central composite design which combines a two-level factorial design with some axial and center points. It is composed of three different parts: a cubic part, which is full factorial, to estimate the linear and interaction terms; another part with axial points to determine quadratic terms, in which central points are defined for all factors except one that assumes its maximum value; and a third part that considers one central point [22]. These points can be further used to screen and evaluate the impact of these five main factors and their two-term interaction on output results.

The variation ranges for input parameters and the design matrix are shown in Tables 3 and 4, respectively. Each parameter value is presented by a coded level. For this three-level design, the parameters levels are shown as [-, 0, +]. Forty-three laboratory measurements were proposed by the three-level central composite design module

of JMP statistical software. In this case, 32 runs were determined by a full factorial design of all 5 parameters, 10 runs corresponded to axial points and one run was the center point (see Table 4).

The gel particles range in size was quantified by using 150, 200, 250 and 325 standard meshes, which are specified in Table 5. Minimum gel particle size was obtained by crushing dried PPGs and then separating gel powders between 250 and 325-meshes. Maximum gel particle size was also obtained by separating gel powders between 150 and 200-meshes. Then, the minimum and maximum gel particle sizes were obtained by averaging opening size values, i.e. 53.5 and 89.5 micrometers. Due to the limitations of commercial sieve mesh dimensions, we could not have a PPG size of 71.5 (average of 53.5 and 89.5) micrometers for the center point of the gel particle size parameter, therefore, we selected the 200 and 250-meshes for obtaining gel particles with dimensions of 68.5 (average of 63 and 74) micrometers for this purpose.

The same issue was highlighted for tube inside radius, where we had three tubes with a regular outer radius of 0.794, 1.588 and 3.175 millimeters but an irregular inner radius of 0.32, 0.52 and 2.21 millimeters. Consequently, we chose the second radius (and not an average value of min. and max.) as the center point of this factor.

RESULTS AND DISCISSION

All experiments in **Table 4** were run by the experimental approach of viscosity test explained in "Experimental approach" section. In four experiments (2, 11, 28, 37), we could not get a stable plateau for pressure drop records, in which, PPGs should be pumped with minimum injection velocity and maximum temperature and therefore their results were rejected. This was due to intrinsic water vaporization of PPGs during tests. Then, we used the least squares method to fit a surface as a quadratic polynomial with main parameters and cross interaction terms.

Table 4: Matrix of runs for response surface design.

Run	Pattern	Gel Particle		Tube inside		Temperature (K)	Pressure	Resistivity	Viscosity
		size (µm)	(Molality of NaCl)	` '	* `	• ` ` `	Drop (Pa)	Factor	(cp)
2	+	53.5 53.5	0	0.32	0.04	298.15 368.15	869429.2	5213	5677
3	+-	53.5	0	0.32	7.0	298.15	1337583	45.8	49.9
4	++	53.5	0	0.32	7.0	368.15	88942.4	9.6	3.3
5	+	53.5	0	2.21	0.04	298.15	109626.7	31394.3	34188.4
6	+-+	53.5	0	2.21	0.04	368.15	13789.52	12519.6	4330.5
7	++-	53.5	0	2.21	7.0	298.15	204084.9	334.9	364.7
8	+++	53.5	0	2.21	7.0	368.15	25510.61	131.8	45.6
9	-0000	53.5	1.75	0.52	3.52	330.65	169611.1	44	28.6
10	-+	53.5	3.5	0.32	0.04	298.15	1087304	4938.9	7097.2
11	-++	53.5	3.5	0.32	0.04	368.15	-	-	-
12	-+-+-	53.5	3.5	0.32	7.0	298.15	1683011	43.7	62.8
13	-+-++	53.5	3.5	0.32	7.0	368.15	238558.7	19.6	8.9
14	-++	53.5	3.5	2.21	0.04	298.15	160647.9	34955.3	50230.8
15	-++-+	53.5	3.5	2.21	0.04	368.15	18615.85	12585.6	5708.8
16	-+++-	53.5	3.5	2.21	7.0	298.15	276479.9	343.2	493.2
17	-++++	53.5	3.5	2.21	7.0	368.15	34473.8	135.2	61.3
18	0-000	68.5	0	0.52	3.52	330.65	88252.93	26.9	14.9
19	00-00	68.5	1.75	0.32	3.52	330.65	310264.2	31.1	20.3
20	000-0	68.5	1.75	0.52	0.04	330.65	120658.3	3134	2038
21	0000-	68.5	1.75	0.52	3.52	298.15	648107.4	95.4	109.5
22	00000	68.5	1.75	0.52	3.52	330.65	158579.5	41.2	26.8
23	0000+	68.5	1.75	0.52	3.52	368.15	75842.36	35	12.8
24	000+0	68.5	1.75	0.52	7.0	330.65	193053.3	28.7	18.6
25	00+00	68.5	1.75	2.21	3.52	330.65	48263.32	231.9	150.8
26	0+000	68.5	3.5	0.52	3.52	330.65	165474.2	34.6	28
27	+	89.5	0	0.32	0.04	298.15	457122.6	3009	2984.9
28	++	89.5	0	0.32	0.04	368.15	-	-	-
29	++-	89.5	0	0.32	7.0	298.15	760492	28.6	28.4
30	+++	89.5	0	0.32	7.0	368.15	110316.2	13.5	4.1
31	+-+	89.5	0	2.21	0.04	298.15	62052.84	19492.8	19135.4
32	+-+-+	89.5	0	2.21	0.04	368.15	11031.62	10986.8	3305.2
33	+++-	89.5	0	2.21	7.0	298.15	112384.6	202.5	198.8
34	+-+++	89.5	0	2.21	7.0	368.15	16547.42	99.1	29.8
35	+0000	89.5	1.75	0.52	3.52	330.65	150995.2	39.1	25.5
36	++	89.5	3.5	0.32	0.04	298.15	936308.4	4253	6111.5
37	+++	89.5	3.5	0.32	0.04	368.15	-	-	-
38	++-+-	89.5	3.5	0.32	7.0	298.15	1449279	37.6	54.1
39	++++	89.5	3.5	0.32	7.0	368.15	205463.8	16.9	7.7
40	+++	89.5	3.5	2.21	0.04	298.15	144100.5	31327.9	45018.2
41	+++-+	89.5	3.5	2.21	0.04	368.15	16547.42	11279.6	5116.4
42	++++-	89.5	3.5	2.21	7.0	298.15	247521.9	307.6	442
43	+++++	89.5	3.5	2.21	7.0	368.15	31026.42	121.2	55

Table 5: Mesh scale.

Mesh	Opening Size (µm)	Average opening Size (μm)
150	105	89.5
200	74	69.3
250	63	52.5
325	44	53.5

Table 6: Primary Model summary of fit.

Model	R ²	$R^2_{ m adj}$
Log (Viscosity)	0.999	0.998

Table 7: Primary Model analysis of variance.

Model	Source	DF	SS	MS	F ratio	Prob> F
	Model	20	306.57999	15.3290		
Log (Viscosity)	Error	18	0.30528	0.0170	903.8451	< 0.0001
	C. Total	38	306.88526			

The least squares method constructs a mathematical function, such as polynomials, which minimizes the sum of the squared residuals between the predicted and actual values. Finally, a primary model (logarithm of viscosity) with statistical characteristics shown in Tables 6 and 7 was obtained.

F-ratio method was also done to test the hypothesis that all the regression parameters (except the intercept) were zero (see Table 7). This was desired since the goal was to declare whether terms in the model were significant. The selection of the most influential parameters was done based on the model. The parameters coefficients used to fit the model have shown their statistical significance. These coefficients values are highly dependent on the scale of the parameters, and since we are interested in the effects size, we needed to examine them in a more scale-invariant fashion. This meant converting parameters from an arbitrary scale to a meaningful one. This was done by transforming the range low to high into -1 to 1. Additionally, these coefficients should be uncorrelated and have equal variances. However, some matrix elements of the coefficients correlation were greater than 0.3. Therefore, the orthogonalized coefficients were computed by premultiplying the column vector of the original coefficients by the Cholesky root of correlation matrix to make them uncorrelated and have equal variances [23].

The effects are sorted by the orthogonalized coefficients in Fig. 3, showing the most significant effects

at the top. A T-test was also done for the hypothesis that each parameter is zero. Probabilities less than 0.05 were considered as significant evidence that the parameter was not zero and it was effective (red color).

As it is marked, PPGs viscosity is primarily influenced by injection velocity (see Fig. 3). It is because the viscosity of fully swollen PPGs is shear rate dependent and PPGs exhibit shear thinning behavior and follow a power law model [13]. Therefore, the viscosity decreases when the injection velocity (flow shear rate) increases. Tube internal radius is the second important parameter. Increasing the radius is equivalent to increasing the cross sectional area and hence decreasing flow shear rate. This means that increasing tube radius will cause the viscosity to increase. Moreover, the temperature has an impact on PPGs viscosity. Since the temperature increment causes the velocity of individual polymer chains to increase, their intermolecular force to decrease and necessarily appeared to decrease the bulk viscosity. This phenomenon is particularly apparent in the case of high temperatures. PPG size and water salinity are also important parameters. There is an inverse relationship between viscosity and particle size. Suspension of smaller particles owing to the greater number of particleparticle contacts per unit volume and hence higher viscosity. Moreover, high salinity brine used to prepare the PPGs

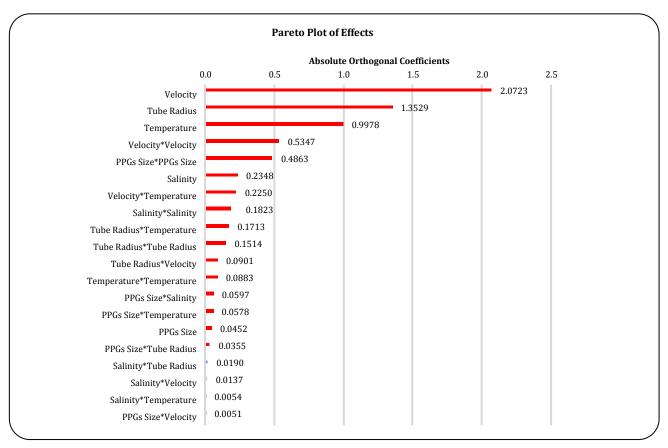


Fig. 3: Pareto Plot of the Effect of Different Parameters.

results in a higher swollen particle strength and accordingly higher resistance factor and viscosity [12].

A proxy model of the logarithm of viscosity was also constructed based on the most important terms screened here. The fitting quality of the model is presented in Table 8. The actual versus predicted viscosity is shown in Fig. 4.

$$\begin{bmatrix} 1.346 \times 10^{-4} \times (D - 71.5)^2 - 0.157 \times (r - 1.265)^2 \\ -0.082 \times (S - 1.75)^2 + 2.507 \times 10^{-4} \times (T - 330.65)^2 + \\ 0.165 \times (V - 3.52)^2 \\ -1.503 \times 10^{-3} \times (D - 71.5)(r - 1.265) + \\ 2.235 \times 10^{-3} \times (D - 71.5)(S - 1.75) \\ +8.978 \times 10^{-5} \times (D - 71.5)(T - 330.65) - \\ 6.598 \times 10^{-3} \times (D - 71.5) \\ +7.051 \times 10^{-4} \times (r - 1.265)(T - 330.65) + \\ 9.448 \times 10^{-3} \times (r - 1.265)(V - 3.52) \\ +1.064 \times (r - 1.265) + 0.153 \times (S - 1.75) + \\ 8.317 \times 10^{-6} \times (T - 330.65)(V - 3.52) \\ -0.027 \times (T - 330.65) - 0.667 \times (V - 3.52) + 4.135 \end{bmatrix}$$

CONCLUSIONS

- 1. Viscosity of these novel PPGs in tubes primarily depended on the injection velocity, tube internal radius, temperature, PPG size, salinity and their two-level interactions.
- 2. The PPG viscosity clearly decreased as injection velocity, PPG size and reservoir temperature increased, also viscosity increased as the tube radius and salinity increased.
- 3. PPG viscosity could be predicted by a simple mathematical model based on different parameters including injection velocity, tube internal radius, temperature, PPG size and salinity.
- 4. Since the injection velocity near the wellbore and reservoir temperature are usually high enough to lower the PPGs viscosity, penetrating deeply into reservoir will be possible.

Nomenclature

D	PPG diameter, µm
k	Permeability, μm ²
L	Length of the tube, cm
r	Internal radius of the tube, mm

Table8: Viscosity model summary of fit.

Model	\mathbb{R}^2	R^2_{adj}	
Log (Viscosity)	0.999	0.998	

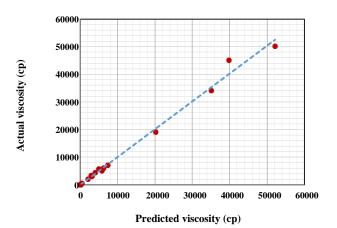


Fig. 4: Plot of Actual vs. Predicted Viscosity Model.

RF	Resistivity factor, -
T	Temperature, K
V	Velocity, mm/s
S	Salinity, molality
ΔP	Pressure drop, Pa
μ	Viscosity of liquid, mPl or cp

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