

An Insight Into The Water Shut-Off Application Of Nano-Sized Preformed Particle Gel Preflush In Heterogeneous Layers

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Abstract

Preformed particle gels (PPGs) are utilized to improve conformance control in mature and water flooded reservoirs to enhance oil recovery and reduce excess water production. In fact, PPG lowers the permeability of totally swept high permeable layers, and thus paves the way for water to enter low permeable and unswept ones. In this study, nine different porous medias with the permeability of around 27 md and porosity of 40% were constructed. Three of them were flooded by nano-sized PPG aqueous phases in different flow rates. Results showed that injecting only nano-sized PPGs does not remarkably reduce the magnitude of permeability. Furthermore, flow-rate had no considerable effect on the efficiency of nano-sized PPG injection process. Three other porous medias were flooded by micro-sized particles. It was obtained that these particles reduced the magnitude of permeability to 7% of its original value, and flow-rate augmentation adversely affects the flooding efficiency.

Finally, the last three porous medias were flooded by a scenario, in which nano-sized PPGs were used as the preflush fluid, and then the porous medias were flooded by micro-sized ones as the main flush. It was obtained that this scenario reduces the permeability of the porous media by 98% and is more appropriate than the single-size flooding process to layer water-shut off. It was because of the fact that porous medias constructed in these experiments consisted of heterogeneous particles with diverse size. Therefore, the size of pore throats was different from each other, which demands using particles with various sizes for permeability reduction.

Keywords: Preformed Particle Gel, Water shut off, Heterogeneous layers, Permeability reduction

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Depletion of hydrocarbon reservoirs and increasing the worldwide demand for energy drives petroleum upstream companies to seek new methods to produce as much oil as possible from reservoirs [20,15,8,19,39]. Water-flooding is the first and the most common approach to increase recovery factor from oil reservoirs [18,29,38,16]. However, excess water production in target wells resulting from water-flooding operations has become a serious problem [18,14,5]. Excessive water production results in occurring numerous problems such as corrosion of production facilities [35,36], high scale precipitation [7,13], imposing an extra load on fluid processing facilities [4], and making some environmental issues [22]. Subsequently, in many cases, oil-producing zones are abandoned due to excessive water production problem, while a remarkable amount of hydrocarbons remain unswept in the reservoir [4,21]. Therefore, researchers have put a special focus on controlling excessive water production from oil reservoirs. Diversity of the geological features of oil reservoirs is the most important issue causing to low oil recovery factor and early excess water production [11,34,10].

Recently, the cross-linked polymer gel has been introduced as a technology to control conformance [32,1,27,28]. However, this technique has several disadvantages such as uncontrollable gelation time, yields disparity due to shear degradation, and change in gelant composition because of adsorption [25,26]. Unlike cross-linked polymers, preformed particle gel (PPG) technology have none of those limitations [11,12,23]. PPGs are powdered polymeric materials possessing elastic features [30]. Furthermore, they are highly hydrophilic, and thus, swell in an aqueous brine solution [2]. PPGs advantages over conventional gelant injection technique have convinced numerous companies to implement this approach, which has had very interesting and satisfying results [24]. Excessive water production has been minimized through PPG injection operations in more than 4000 wells in China [4] and several wells in Iran.

Potential and advantages of PPG injection operation could be attributed to the interaction between gel and formation particles [23]. In a PPG injection scenario, depth of penetration of particle gels can be determined through injected liquid volume and PPGs' concentration data [25,26]. However, poor designing of PPGs size results in faster pressure growth, and thus, particles cannot penetrate deep enough in the formation [24]. Saghafi et al. conducted several experiments to study the penetration of gel particles in various porous medias. The results show that PPGs can move forward through smaller pore throats under the mechanism of deformation. In this intend, it was reported that when the size ratio of particles to pore throats of the porous media becomes lower than a critical value (about 1.9, depending on the chemical composition of PPG), PPGs can be deformed and change their shape to move forward though pore throats. However, when the size ratio exceeds this critical value, PPGs have to be crushed in order to pass through throats, which is unfavorable and results in faster pressure growth [23-26]. Therefore, in such cases the output pressure of injection pumps reach the highest limit after injecting a low volume of particles into the subsurface resulting in a lower depth of penetration and cross-flow occurring [31,3]. Effect of other parameters such as flow rate, PPG concentration, cross-linker concentration [17,14], adsorption, and retention of particles [23] have investigated on the efficiency of PPG injection operations by various researchers. However, the effect of the size distribution of particles on the efficiency of PPG injection has received fewer attentions. Oil reservoirs, especially carbonate ones, are very heterogeneous, and the size of pore throats in their porous medias are very diverse. Pore throats regarding primary porosity are very smaller than the ones resulted from secondary porosity. Therefore, such layers demand being flooded by particles with various sizes.

In this study, an optimized gel sample through the procedure explained by Saghafi et al. [24] was synthesized. Then, the prepared gel was grounded to achieve nano and micro-sized PPGs. The efficiency of nano-sized and micro-sized PPGs on permeability reduction of target porous medias was determined experimentally and then compared with each other. Also, the effect of flow rate was determined in those experiments. Furthermore, a scenario in which nano-sized PPGs used as the preflush fluid and micro-sized ones were the main flush was studied.

Materials And Methods

Materials

Synthesized PPG

The material utilized to synthesize particle gels is listed in Table 1. N,N'-methylenebis (Acrylamide) was the linking agent [17] in the synthesis process, which was purchased in Beijing Chemicals. N,N',N,N'tetramethylethylenediamine was used as the catalyst of the process [9], and sodium persulfate was the reaction initiator[9]. They both were purchased from Beijing Chemicals company too. Sodium montmorillonite, which was utilized to enhance the mechanical strength of the synthesized gel [35], was purchased from Sigma Aldrich company. Furthermore, high pure (99.99%) nitrogen gas needed for nitrogen tapping process was provided from the Delvar Afzar industrial group.

Table 1: Materials utilized to synthesize PPGs

Function	Monomers		Crosslinker	Mechanical strength	TSA	Initiator	Catalyst	
					enhancement			
Material	AM	VP	AMPSNa	MBA	SM	TSA	APS	TEMED
Wt %	5.6	9.0	12.9	0.57	1.77	0.10	0.10	0.05

Materials displayed in Table 1 were implemented in PPG synthesize procedure. For the first step, 1.77 gr sodium montmorillonite (SM) was poured to a 70 mL deionized water sample. The aqueous phase was stirred and then ultrasonicated to attain a homogenous liquid. Afterwards, 5.6 gr acrylamide (AM) and 9 gr vinyl propylene (VP) were added to the aqueous phase, respectively. Then, 12.9 gr 2-acrylamide-2-methylpropane sulfonic sodium salt (AMPSNa) in addition with 0.1 gr thermal stabilizing agent (TSA) were poured to the aqueous phase and stirred for 60 minutes to disperse all monomers. At this point, nitrogen tapping process was started to prevent reaction between oxygen and methylenebis acrylamides (MBA), which is added at next.

In the next step, after achieving a homogenous monomers dispersion, 0.57 gr methylenebis acrylamide was added to the aqueous phase as the crosslinking agent and stirred for 40 minutes. Afterwards, 0.1 gr initiator and 0.05 gr catalyst agent were added to the mixture. Nitrogen tapping was stopped at this point. After 30 minutes, the synthesized hydrogel was formed through an exothermic reaction. The synthesized hydrogel was immersed in deionized water for 24 hours at room temperature, to make sure that the reaction moved forward completely. Then, the hydrogel was cut into smaller pieces and put in the oven for 24 hours at 55°C in the vacuumed condition. The dried product, which is shown in Figure 1, was the PPGs utilized in this study.



Figure 1: Dried hydrogels utilized as PPG in this study

Crushed carbonate rocks

Slim tube

Several reservoir core plugs provided by National Iranian Oil Company (NIOC) were washed and cleaned by a deanstark apparatus by means of the toluene-methanol mixture at 90°C for two weeks. Then, the core plugs were grounded by a Jaw crusher. Subsequently, crushed samples were sieved into three fractions using meshes No. 270, 325, and 400. The size of particles varied from 20 to 53 micrometre. Then they were packed into the slim tubes to simulate heterogeneity of carbonate rocks. Stainless steel slim tubes with a length of 51 cm were utilized in this study. The slim tube was packed through crushed and sieved rock samples. The permeability of all tubes (9 numbers) prior to PPG injection was obtained to be around 27 md.

Experimental apparatus

The apparatus illustrated in Figure 2 was utilized in displacement experiments in this study. The set-up consisted of slim tubes with a length of 51 cm, a Vinci positive displacement pump, a recombine cell to homogenize PPG aqueous phase, oven, several pressure transducers, and data acquisition system.



Figure 2: Experimental set-up utilized in this study

Experimental And Methodology

Flooding test

Prepared slim tubes were flooded by various aqueous phases to evaluate the performance of synthesized PPGs (both micro and nano-sized) in permeability reduction of the porous media. Resistance factor (RF) and a residual resistance factor (RRF) were calculated as indicators of the PPG flooding performance. This experiment was conducted through the following steps.

1- The packed slim tube was vacuumed and then saturated with deionized water. The pore volume and porosity of the porous media were calculated through slim tube weighting before and after saturation, which is written in Equations 1 and 2.

1

$$PV = \frac{W_s - W_d}{\rho_w} \qquad \text{Eq.}$$

$$\varphi = \frac{PV}{V_t}$$
 Eq. 2

Where PV refers to the magnitude of pore volume in mL, W_{s_s} and W_d represent the weight of saturated and dry porous media in gr, respectively. ρ_w denotes to the density of water at the desired temperature. φ is the porosity of the porous media. V, is the total volume of the porous media.

2- The slim tube was flooded by deionized water in various flow rates, ranging from 0.5 to 10 mL/minute to calculate the permeability of the porous media. According to Equation 3, by plotting the magnitude of pressure drop along with the porous media versus flow rate permeability can be determined.

$$\Delta P = \frac{q\,\mu L}{KA} \qquad \text{Eq. 3}$$

Where, ΔP is the pressure difference between inlet and outlet of the porous media in psi. q refers to the magnitude of flow rate in mL/minute. μ (cp) is the viscosity of the deionized water. L and A represent the length and cross-sectional area of the porous media in cm and cm², respectively. K is the permeability of the porous media in Darcy.

3- The porous media was flooded by several PVs of brine. In this step, the porous media was saturated with brine, and the magnitude of pressure drop was recorded for brine injection at different flow rates (ΔP_i)

4- Brine-saturated porous media was flooded by fully swollen PPG aqueous phase at various flow-rates. Same as brine injection, the pressure drop was recorded in this step too (ΔP_n)

5- Secondary brine injection was conducted on PPG bearing porous media. The higher the secondary brine injection pressure (ΔP_S), the better the performance of the PPGs in the reduction of the permeability of the porous media.

Resistance Factor (RF) Calculation

Resistance factor (RF), which is effective viscosity of PPG aqueous phase in the porous media, is defined as the ration of the pressure drop in PPG injection to the primary brine injection process. RF is highly related to the retention (entrapment plus adsorption) of PPGs in the porous media. This parameter was calculated at each flow-rates. The equation by which the magnitude of RF was calculated is written below.

$$RF = \frac{\Delta P_p}{\Delta P_i}$$
 Eq. 4

Residual Resistance Factor (RRF) Determination

Residual resistance factor (RRF) is an important parameter indicating the permeability reduction of the porous media after PPG flooding process. This parameter is calculated to ensure that an acceptable portion of injected PPGs remains in the porous media, and the majority of pore throats are plugged by particles. Equation 5 calculates RRF parameter.

$$RRF = \frac{\Delta P_s}{\Delta P_i}$$
 Eq. 5

Results And Discussion

Nano-sized PPG flooding

Prepared PPGs were grounded and milled using a planetary ball mill apparatus to produce nano-sized PPGs. Then the grounded PPGs were gradually added to the brine water sample and stirred for 24 hours to achieve a homogeneous PPG dispersion with a particle concentration of 3000 ppm. Afterwards, the colloidal properties of the fully swollen nano-sized PPGs aqueous phase such as average particles size and zeta potential were measured through Malven zen 3600 zetasizer at 28ºC. The magnitude of average zeta potential for these particles was obtained -14 my, which indicates that there is a slight repulsive force between particles that can prevent immediate agglomeration of them. Figure 3 illustrates the size distribution of particles in brine water. As well depicted, the size of swollen particles dispersed in the aqueous phase ranged from 220 to 380 nm, with the average size of 345 nm. Both zeta potential and particle size measurement results indicate that nano-sized PPGs are dispersed homogeneously and without agglomeration in the brine water sample.



Figure 3: The size distribution of nano-sized PPG in brine sample after stirring for 24h

Three different porous medias through slim tube packing process were constructed. Size of crushed cores utilized in this experiment varied from 20 to 52 μ m. Characteristic of these slim tubes are displayed in Table 2. All slim tubes mentioned in

Slim tube No.	Porosity	Permeability (md)	Pore volume (mL)
1	40.05	27.1	7.82
2	40.1	27	7.87
3	40.07	27	7.85

 Table 2: Characteristics of slim tubes used for nano-sized PPG injection

Table 2 was flooded by brine, nano-sized PPG fluid, and brine consecutively under flow rates of 0.05, 0.07, and 0.1 mL/ min, respectively. Pressure data obtained from PPG injection

processes were recorded versus the volume of injected PPGs, which is shown in Figure 4. Important parameters such as RF, RRF, and residual permeability for each slim tubes are calculated and displayed in Table 3.

Tube No.	q (mL/min)	ΔP_{i} (psi)	ΔP_p (psi)	ΔP_{S} (psi)	RF	RRF	K _{residual} (md)
1	0.05	58	440	131	7.586	2.258	11.95
2	0.07	81	584	180	7.209	2.222	12.15
3	0.1	117	820	259	7.008	2.205	12.19

Table 3: Data obtained from the PPG flooding process of slim tubes

Based on data presented in Table 3, PPG injection rate doesn't have a significant effect on RRF and residual permeability of the porous media. However, a slight reduction in RRF by flow-rate augmentation is because of shear force increment imposed from aqueous phase movement to particles. Furthermore, it is obvious that nano-sized PPGs cannot reduce residual permeability remarkably. It can be explained by the fact that smaller particles move forward through bigger pore throats (representative of secondary porosity) without any resistance. Consequently, only smaller pore throats (primary porosity) are plugged by PPG particles. Pressure drop data of PPG injection processes versus injected volume are shown in figure 3. The magnitude of pressure drop in stabilized flow increases with flow-rate, which is justified by darcy law, in which pressure drop is directly related to the flow-rate. The first picks observed in Figure 4 may be resulted from agglomerations of nano-sized PPGs in larger pore throats, which are not stable and are eliminated by further injection. The injection process in these experiments was continued until pressure remains constant for several injected pore volumes.



Figure 4: Pressure drop data versus injected PPG PV in various flow rates

Micro-sized PPG flooding

As illustrated in Figure 4, nano-sized PPGs agglomerations last longer and require a higher pressure to break down in higher flow-rates. It can be justified by the fact that in higher flow-rates larger amount of particles enter the porous media suddenly when the flooding process starts. Therefore, in such conditions, particles form more stabilized agglomerations in pore throats, demanding higher pressure to break down. Furthermore, the required volume of PPG aqueous phase to achieve stabilized pressure increases by flow-rate, which results from agitation and turbulency increment in higher flow-rates.

Micro-sized PPG injection was conducted to make a comparison with results obtained from nano-sized one. To do this, synthesized PPGs were grounded using a disk mill to produce micro-sized particles. Then, grounded particles were poured into the brine sample and stirred to achieve swollen and fully dispersed aqueous phase. The average size of swollen particles in brine was 149 μ m. Afterwards, flooding procedure was conducted the same as nano-sized PPGs injection. Results obtained from this experiment are displayed in Table 4.

Tube No.	K	q (mL/min)	ΔP_i (psi)	ΔP_p (psi)	ΔP_{S} (psi)	RF	RRF	K _{residual} (md)
4	27.1	0.05	56	3574	995	63.82	17.76	1.52
5	26.95	0.07	85	3902	1023	45.9	12.03	2.23
6	27	0.1	112	4442	1299	39.66	11.59	2.32

Table 4: Results obtained from micro-sized PPG injection

Results shown in table 4 disclose that the magnitude of RF decreases with flow-rate increment. It is because of the shearthing behavior of the PPG aqueous phase [6,37]. The viscosity of the PPG liquid decreases at higher shear rates, which lowers the magnitude of viscous force against the flow. Furthermore, It was obtained that RRF behaves like RF and have an opposite relation with flow-rate. It can be justified by the fact that higher shear rate and velocity supply the required energy for PPG deformation, and thus lowers the magnitude of particles entrapped in the porous media. Figure 5 illustrates injection pressure versus injected pore volume in various flow-rates. The peeks observed in nano-sized PPGs are not detected in this experiment. Because larger particles don't have a remarkable tendency for being agglomerated. Therefore, it can be concluded that unlike nano-sized PPGs, micro-sized ones are not agglomerated in the porous media.



Figure 5: micro-sized PPG injection pressure versus injected PV in various flow rates

As depicted in Figure 5, pressure increases sooner and faster when PPGs flooded by the flow-rate of 0.05 mL/min. Furthermore, in this flow-rate, the stabilized flow is achieved after injecting a low volume of PPGs into the slim-tube, while it's RRF value is higher (See table 3). It shows that in laminar flows particles have more chance to be entrapped in the porous media, and thus the permeability reduces higher and faster.

Hybrid-sized PPG injection

Three other heterogeneous porous medias were constructed via the procedure explained previously. Porosity and permeability of slim tubes were measured in the laboratory, which was so close to the characteristics of previous ones. Table 5 illustrates data obtained from slim tubes characterization experiments. Slim tubes were flooded under different flow-rates of brine, nano-sized PPG, micro-sized one, and secondary brine, consecutively. PPG injection parameters are measured and calculated for these slime tubes, which are displayed in table 6.

Slim tube No.	Porosity	Permeability (md)	Pore volume (mL)
7	40	27	7.81
3	40	27.1	7.81
)	39.8	27	7.77

Table 5: characteristics of constructed slim tubes utilized in this experiment

		,	1	1	,	1		
Tube No.	q (mL/min)	ΔP_i (psi)	ΔP_p (nano)	ΔP_p (micro)	$\Delta P_{S} ({ m psi})$	RF	RRF	K _r (md)
7	0.05	60	444	5898	2018	98.3	33.63	0.8
8	0.07	84	587	6271	2375	74.65	28.27	0.95
9	0.1	120	827	6799	2756	56.65	22.96	1.17

Table 6: PPG injection parameters obtained from hybrid sized PPG injection process

Porosity, permeability, and the magnitude of pressure drop in primary brine injection were very close to their values in previous experiments. Therefore, It can be concluded that the process by which slim tubes have been constructed is highly repeatable. Furthermore, the similarity of pressure drop measured in tubes 7 to 9 with tubes 1 to 3 is concrete evidence to testify the repeatability of PPG injection process. The magnitude of RF decreases by flow-rate, which shows that PPG aqueous phase has a shear-thinning behavior. By comparing RRF results obtained from this experiment with previous ones, it can be concluded that conducting nono-sized PPG preflush prior to micro-sized PPG flooding operation is helpful in reducing the permeability of aimed zone. Nano-sized particles plug smaller pore throats, which are inaccessible for micro-sized PPGs. Subsequently, conducting micro-sized PPG flooding after nano-preflush reduces the magnitude of permeability more than the case without preflush. In oil reservoirs, especially carbonate ones, Pores throat sizes are very diverse, ranging from nanometer to hundreds of micrometre. Thus, preflushing the layer with smaller sizes prior to the main aqueous phase can be very helpful in permeability reduction of the layer.

Conclusion

The potential water shut-off application of optimized nano-sized PPGs was investigated in this study. The following conclusions are obtained.

1- Nano-sized PPGs are dispersed homogeneously in brine aqueous solution via a slight electrical repulsive force existing between particles.

2- Nano-sized PPGs are agglomerated in the porous media and block larger pore throats resulting to pressure increment. However, blocks and agglomerates are eliminated by further fluid injection and the injection pressure drops at that point.

3- Nano-sized PPGs are only capable of plugging small pore throats, which are inaccessible for micro-sized ones.

4- Nano-sized PPGs reduced the permeability of the porous medias by 45%, however, this value for micro-size ones was 93%. This happened because of the fact that nano-sized particles couldn't entrapped behind larger pore throats and passed through them.

5- Increasing PPG injection flow-rate adversely affects the efficiency of PPG injection process. Because it prevents particle entrapment in the porous media, and thus, the permeability of the porous media does not decrease significantly.

this scenario decreases the permeability of the porous medias by 98%. Because either small or large pore throats are blocked in this scenario.

6- Finally, porous medias were preflushed by nano-sized particles and then flooded by micro-sized ones. Results showed that

Nomenclature

ϕ	Porosity (%)
ΔP	Pressure Drop (psi)
ΔP_i	Pressure Drop of Initial Water Flood (psi)
ΔP_p	Pressure Drop of PPG Injection (psi)
ΔP_s	Pressure Drop of Secondary Water Flood (psi)
μ	Viscosity (cp)
К	Permeability (md)
K _r	Residual Permeability (md)
PPG	Preformed Particle Gel
PV	Pore Volume (number)
q	Flow-rate (mL/minute)
RF	Resistance Factor
RRF	Residual Resistance Factor

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