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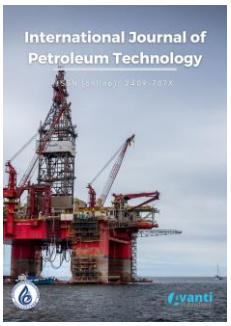


# Overview of In-Situ Gelation Behavior of Gel Systems in Porous Media

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## ABSTRACT

Cross-linked polymer gel has been widely applied in profile control and water plugging due to its effective cost, wide suitability, excellent performance, and flexible gelation time. Previous research mainly focused on the bottle tests, reaction kinetics, and rheological properties of the gels, but the works of literature about the in situ gelations of gel placement in the porous media are relatively few. The study of the in-situ gelation behavior of gel systems is widely summarized in this paper, and the research tendency is proposed. The important practical questions, including the accurate lateral distance of gel placement, the variation of gel properties, and the injection pressure profile in the process of gel injection, should be resolved by laboratory and numerical research to enhance gel treatment success rate.

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# 1. Introduction

During the water flooding and polymer flooding process, the volumetric efficiency is poor because of the severe heterogeneity of reservoirs. Now cross-linked polymer gel has been widely applied in profile control and water plugging due to its effective cost, wide suitability, excellent performance, unique structure, and flexible gelation time. Previous research mainly focused on the effect of concentration and type of crosslinker, concentration and type of polymer, temperature, pH value, and salinity on the gelation time and gel strength in the bottle test [1], reaction kinetics [2] related to the gelation process and rheological properties of the gels [3,4]. In contrast, there are relatively few works of literature about the in-situ gelation and the accurate lateral distance of gel placement in the porous media. However, earlier research [3,4] indicated that the shear rate could alter the gelation time during the gelation process. Thus, the in-situ gelation mechanism, including the in-situ gelation time, the injection pressure profile, and the accurate lateral distance of gel placement which are critical parameters for the success of oilfield operation, must be studied urgently.

# 2. Overview

## 2.1. Experimental Research on the In-Situ Gelation Behavior of Gel Systems

According to the different kinds of gel systems and porous media, the prior works mainly focused on the gelation behavior of polyacrylamide/chromium(VI)/thiourea, xanthan/Cr(III), acetate, glycolate, and malonate complexes of Cr(III)/polyacrylamide gel systems, phenol-formaldehyde/polymer gel systems, polyacrylamide/ aluminum citrate colloidal-dispersion gel (CDG) system, sodium silicate gel and bio-polymer gel system(controlled by pH) in sand packs, sandstone cores or the fractures.

### *2.1.1. Polyacrylamide/ Chromium(VI)/Thiourea Gel System*

Huang *et al.* [5] used a sand pack holder 12 inches (30.5cm) long and 1.5-inch Internal Diameter(ID) to study the in-situ gelation of a polyacrylamide/chromium(VI)/thiourea gel system. In the experiments, the in-situ gelation time in unconsolidated sand packs was much shorter than the gelation time in the bottle test, affected by the in-situ shear rate, and had a minimal relationship with sand particle size. When gelation occurred before 1 PV of gel solution was injected, the viscosity of the gel solution increased as the distance from the point of injection increased. The study also found that the contaminants of brass fittings and PVC would accelerate gelation time. McCool *et al.* [6] and Marty *et al.* [7] studied the mechanisms of in-situ gelation for a polyacrylamide/chromium(VI)/ thiourea gel system in 4-ft-long sand packs. During the injection process, a localized high-flow-resistance region developed well behind the leading edge of the gel-solution bank and ultimately caused the sand pack to plug. It was hypothesized that gel aggregates were formed and retained in the porous matrix. The existence of these aggregates was established by experiments using equilibrium dialysis. McCool *et al*. [6] postulated that "aggregates were filtered out of the flowing solution by interactions with the porous matrix, by reacting with previously retained polymer and by selective straining of the larger aggregates that could not pass-through pore constrictions."

### *2.1.2. The Xanthan/Cr(III) Gel System*

Jousset *et al.* [8] focused on the effect of high and medium shear rates on the mechanism of in situ gelations during the gel solution injection period. The experiment used a stainless steel slim tube of 15 ft length and 0.295 ID composed of five sections to simulate the flow conditions encountered in the vicinity of a wellbore during a xanthan /Cr(III) gel treatment. The experiments indicated that in situ gelations in the porous medium began at a point where the viscosity of this system started to increase in the bottle test. During the in-situ gelation, a narrow region of high flow resistance developed at relatively low velocities, and its location is a linear function of frontal advance rate. The magnitude of the developed resistance correlates with frontal velocity, but when the advance rates were 83-118ft/d, high flow resistance did not develop. Hejri *et al.* [9] revealed the mechanism of permeability reduction in situ gelations of a xanthan/Cr(III) gel system in unconsolidated sand packs at frontal velocities between 3 and 120 ft/D. The experiments used two different types of sand packs; one was a 4.5-ft-long, 1.44-in-ID

Lucite tube, and the second one was 15-ft-long, 0.295-in-ID stainless-steel tube. Their research revealed that permeability reduction in the sand pack began at a location where the gel solution residence time was the same as when the system viscosity increased in a bottle test. The primary mechanism of flow resistance development during continuous gel-solution injection was the retention of gel aggregates. The location of the high-flow-resistance region was a function of frontal-advance rate, and the developing rate of the high-flow-resistance region was related to in-situ shear rate and permeability. McCool *et al.* [10] researched fluid/rock interactions between Xanthan/ Chromium(III) gel systems prepared with chromium chloride, chromium acetate, and a chromium diamine salt in the dolomite core materials obtained from a San Andres dolomite reservoir in Texas. The experiments revealed that the increase in gal pH value due to fluid/rock interactions resulted in the precipitation of chromium and then brought about incomplete gelation in the dolomite materials. In contrast, the chromium acetate/xanthan gel system formed a very high residual resistance factor in a sand pack at pH values between 4.1 and 4.5. Maddinelli *et al.* [11] established a new approach to characterize the gelling process of Cr(III)/HPAM inside the porous media by using NMR relaxometry and imaging. The results demonstrated that gelation time in porous media was shorter than in bulk solutions due to a mineralogical effect on the pH of the saturating fluid.

### *2.1.3. Other Gel Systems*

Bartosek *et al.* [12] used acetate, glycolate, and malonate complexes of Cr(III)/polyacrylamide to evaluate the stability of Cr(III) complexes in bulk solution and Cr(III) propagation in the sand packs, Clashach sandstone cores, and Berea sandstone cores. It was found that Cr(III) precipitation and Cr(III) retention in porous media were closely related phenomena by comparing bulk solution studies with core flooding, and the gelation delaying ligands added into the gel solutions could significantly enhance the stability of Cr(III) and its propagation in porous media. It was also detected that Cr(III) precipitation could be reversible by adding strong complexing agents, and precipitated Cr(III) was capable of reacting with subsequently injected polymer and then caused a rise in the injection pressure.

Koines *et al.* [13] investigated the effect of the composition of the core material on the gelation process for polyacrylamide/chromium(III)-malonate by using Berea cores, Bentheimer cores, and a North Sea reservoir sandstone. Clay and carbonate content in the core significantly impacted the gelation process. In order to make the gel form successfully in a Berea core, the following conditions should be fulfilled: ① sufficient buffer should be added to keep the pH below 7; ② Sufficient chromium should be injected (more than ten times the concentration that gives gel in bulk) in order to saturate the clay by ion exchange; ③ Oxygen should be removed from all fluids used in the experiment to prevent free radical degradation of the polymer.

Seright [14] examined the effect of gelant viscosity, degree of gelation, and gravity on gel placement in fractured systems by using Cr(III)/acetate/HPAM, resorcinol/formaldehyde, Cr(III)/xanthan, aluminum/citrate/HPAM and other gelants and gels. The results suggested that gelants can penetrate readily into the rock matrix before gelation, but gel propagation is extremely slow or negligible after gelation. Preformed gels improved sweep efficiency much more effectively than gelants that formed gels in situ. During injection of aqueous gelants into fractures, viscous forces usually dominated over gravity forces, and the gelant front's position would not be significantly influenced by gravity.

In the fractures, water leak off from formed Cr(III)-acetate-HPAM gels during extrusion was described by Seright using a new model [15]. The new model successfully obtained the occurrence of wormholes and pressure gradients during gel extrusion through fractures compared to the conventional model. When the width was smaller than 0.04 inches, gel mobilization required pressure gradients similar to those during gel injection, but gels only needed lower-than-expected pressure gradients for mobilization when the fractures were wider (0.08-0.16 inches).

McCool *et al.* [16] injected the in-line-mixed polyacrylamide/chromium acetate gelant into a tube composed of 5 sections with 1,031-ft-long and 0.0566-inch-ID to simulate gel treatment for fractures. The results indicated that steady flow resistances of the downstream sections forming in the injection of in-line-mixed gelant were higher than that forming in the injection of preformed gel. The solvent expelled from the gel due to the syneresis

produced by the deformation during and after gelation lubricated the downstream flow of gelants and preformed gels.

Vossoughi *et al.* [17] studied a newly discovered bio-polymer produced by Cellulomonas Flavigena strain KU in sand packs composed of two and three 10 cm long cells. The experimental data revealed that high permeability reduction could be achieved along the entire sand pack. More importantly, the initial permeability of the sand pack can be restored by injecting sodium hydroxide solution. In addition, the new bio-polymer system provided superior selectivity compared to other available systems.

Albonico *et al.* [18] investigated the phenol-formaldehyde/HPAM gel system, and it was discovered that the phenol and formaldehyde crosslinkers would react to marked turbidity in the gelant solutions well before gelation. Filtration experiments and core floods showed that the presence of phase-separated material has a negligible impact on gelant propagation and injectivity. In addition, the loss of phenol by partitioning into the crude oil with which it comes into contact was identified as an important issue for these gelants. Numerical simulations revealed that the method of a phenol slug injecting into the matrix formation ahead of the gelant might be potentially effective in compensating for poor phenol propagation.

Bryant *et al.* [19] investigated the injectivity of phenol-formaldehyde/polymer gels. The experiment certificated that injectivity loss caused by filtration and viscosification were mingled in field treatments, and the experiments involving multiple PV injection, step changes in flow rate, and in-line capillaries could be used to evaluate them independently in the laboratory. Tests also revealed that filtration of pregel aggregates was the main reason for the mobility reduction of the gelant in sand packs and cores, which was strongly correlated to the age of the gelant but relatively insensitive to the permeability or the presence of residual oil.

Ranganathan *et al.* [20] studied the gelation behavior of a polyacrylamide/aluminum citrate colloidal-dispersion gel (CDG) system in the sand pack with 4-ft-long and 1.5-inch-ID at frontal advance rates of 2 ft/D. The size distribution of aggregates at selected times after mixing was determined by membrane dialysis. The experimental results show that the gel solution flow was similar to polymer solution in the porous media and gel retention occurred at the inlet of sand packs, and aggregates were mainly retained at the interface between high permeability regions and the relatively low permeability regions. The crosslinker was significantly retained in the porous media in the flow process.

Stavland *et al.* [21] researched the gelation process of sodium silicate in the Bentheim sandstone cores with 25-cm-length, 3.8-cm-ID. The work demonstrated that sodium silicate should be diluted in brines with a low divalent ion concentration addressing the need for a sufficiently large preflush. The gelation time, which could be measured by turbidity, was controlled by the silicate concentration and the activator, the temperature, the salinity, and the injection flow velocity.

## 2.2. Numerical Simulation of the In-Situ Gelation Behavior of Gel Systems

### *2.2.1. Lab-Scale Numerical Simulation of the In-Situ Gelation Behavior of Gel Systems*

Hubbard *et al.* [22] conducted two in-situ gelation experiments using xanthan/$CrCl_3$ to study the cross-linking process in the Clashach sandstone cores. In the experiment, chromium was labeled with $^{51}Cr$, which could be detected by the gamma rays to observe in-situ total chromium profiles during the core floods. The convection-dispersion equations, including transport, adsorption, and chemical reactions, were used to simulate the observed behavior in the gelation process. Model C was verified to be correct because it involved a fast first reaction which implied that all the chromium was associated with a relatively small amount of xanthan; thus, the phenomenon that the xanthan without chromium first breaks through the sandstone cores could be explained.

Todd *et al.* [23] established the filtration model to simulate the in-situ gelation of polyacrylamide/chromium(VI)/thiourea system in the linear displacements performed by McCool *et al*. [6] and Marty *et al*. [7]. The results from the simulation model showed that the location of the zone of rapid filtration was determined by gelation kinetics, the maximum treatment depth was limited by the distance where the high-flow-resistance zone developed for the gel system studied, and the location of the region where plugging occurred was a linear function of flow velocity.

After that, Todd *et al*. [24] built up a new redial model which described gelation behavior during the period of gel solution injection and the subsequent shut-in period to simulate practical gel treatment cases. The simulation revealed that the gelling solution would enter each layer at rates proportional to the permeability of the layer in the gel treatment for formations with multiple layers of widely varying permeability; even the lowest permeability layer simulated (0.035 Darcies) was plugged by the gelling solution. So, isolation of high permeability zones is required for in-depth permeability modification.

Khachatoorian *et al.* [25] verified McCool's filtration hypothesis [6] using their simulation results and provided insights into the phenomena of in situ gelations both in petroleum recovery and subsurface heavy metal stabilization. The simulated results were in close agreement with experimental data for the case of polyacrylamide/redox system in sand pack. The viscosity of the polymer in the porous media increased exponentially with time depending upon the position of the sand pack.

Stavland *et al.* [26] established a modified UTCHEM simulation model, including retention, gelation strength, and gelation rate for the in-situ gelation of a xanthan/Cr(III) gel system. In the model, the dissolution rates of carbonates explained that the main way of retaining the crosslinker in Berea cores is precipitation, and pH dominated the gelation process. The results revealed that the gelation process in the porous media could be regarded as the continuous filtration of aggregates. The beginning gelation time in porous media with low carbonate content was similar to the gelation time in bulk experiments but was lower in Berea cores due to the increased pH.

#### *2.2.2. Field-Scale Numerical Simulation of the In-Situ Gelation Behavior of Gel Systems*

Surguchev *et al.* [27] developed a validated gel model to imitate the gelation process of the xanthan/Cr(III) gel system. The new model described the important features, which included improved surfactant phase behavior description, modeling of the thermal effects, chemical reactions including gelation kinetics, gelation strength, retention models, buffering capacity of the reservoir material, extended tracer routine, and a new mufti ion exchange routine. The innovative model proved to be useful in history matching and evaluating treatment effects in several field pilots in the North Sea.

Dang *et al.* [28] successfully investigated the gelation behavior of Cr(III)-acetate-HPAM from laboratory to full-field scales. In experiments, it was found that gelation time and gel strength were the successful keys to the conformance control process, and they strongly depend on the polymer gel composition, temperature, salt type, salt concentration, and pH. The numerical simulation was successfully developed with an innovative history matching approach for the polymer conformance control in the Lower Miocene reservoir of the White Tiger field.

### 2.3. Study on High Temperature and High Salt Resistance of Gel Systems

According to some research [29,30], in addition to commonly used polymer systems such as polyacrylamide (PAM), partially hydrolyzed polyacrylamide (HPAM), and copolymers based on acrylamide monomer (AM) also include other monomers with AM can improve the tolerance for temperature and salinity.

Liu *et al*. [31] developed a gel system formed by the terpolymer (L-1) and a new cross-linking system (HB-1), and its properties were systematically studied in the condition of extremely high temperatures. Studies have shown that the gel system could form stable continuous 3D network structures in high temperatures (120°C–200°C). When the temperature is 200°C, the grid size of the network structure is about 10 μm, but the strength of the gel system can still be maintained at code F-G. When the salinity of the water solution is 200,000 mg/L, the gel system on the contact surface can still maintain a small grid size of 3D network structures (about 10 μm).

According to He *et al*. [32], an environmentally friendly improved inorganic aluminum gel composed of polyaluminum chloride (PAC) as the main agent, urea as an activator, and sodium sulfate as syneresis inhibitor was developed to solve the problems such as application limitations of polymer gels under harsh reservoir conditions. The gel was highly temperature and salt-resistant. It was stable for 120 days under 130℃and could tolerate sodium chloride concentrations up to 150 g/L and calcium chloride concentrations up to 25 g/L. The gel

had a good ability to reduce permeability and could effectively plug high permeability areas. The research showed that the gel had a certain degree of control under harsh reservoir conditions, providing a basis for field application.

## 3. Research Trend of In-Situ Gelation Behavior in the Future

Although much work about the effects on the gelation time and gel strength in the bottle test, reaction kinetics related to the gelation process, and rheological properties of the gels have been done, much more work is needed to cover field applications. In particular, several important practical questions must be solved: What is the accurate lateral distance of gel placement? How does the injection pressure profile vary? Will the front gel which may form a gel, block the subsequent gelants in the injection process of in-depth gel placement? How are gel properties affected by continued gel curing reactions and shear degradation during the in-situ gelation in the porous media? Once these and other practical questions about the in-situ gelation mechanism are answered, the gel treatments used as the permeability modification treatments could obtain a successful guarantee and then improve waterflood sweep efficiency and oil recovery. So, we need to obtain these answers with laboratory and numerical research.

## 4. Conclusion

Through the overview of the prior literature, the gelation time and gel strength in the bottle tests, reaction kinetics, and rheological properties of gel systems have been studied extraordinarily clearly. However, the important practical questions should be solved, including the accurate lateral distance of gel placement, the variation of gel properties, and the injection pressure profile in the process of gel injection with laboratory and numerical research in order to enhance the success rate of gel treatments which are proved as the effective flow control agents to resolve the increasing excessive water production in the mature reservoir.

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