



An experimental study on application of nanoparticles in unconventional gas reservoir CO₂ fracturing

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ABSTRACT

Gas production from low permeability unconventional reservoirs is still a challenge to the world. Although hydraulic fracturing has been successfully applied in unconventional gas production, its limitations are obvious, such as formation damage, water blocking, low stimulating effect and the requirement of sufficient water. To avoid these problems, liquid CO₂ has been pumped as a fracturing fluid into unconventional reservoirs. But as a consequence of low density and viscosity of super critical phase CO₂ in the formation, CO₂ fracturing suffers from low sweep efficiency that manifests as viscous fingering. So various additives have been applied to improve CO₂ fracturing effect. This paper introduces a novel additive, nanoparticles, and presents an experiment to evaluate its effect on CO₂ fracturing.

In this paper, a core flooding experiments was conducted to simulate the fracturing process, in which liquid CO₂ was injected into a core to drainage brine or nanoparticles solution. During the process, CO₂ distribution and pressure drop were real-time measured with a modified medical CT scanner and pressure transducers. A significant difference is observed between with and without nanoparticles. The saturation files show that CO₂ fingering was decreased and the drainage area was improved with the action of nanoparticles. Meanwhile, the CO₂ injecting pressure raised, which implies that nanoparticles could offer higher pore pressure in fracturing. These observations suggest that a nanoparticle-stabilized foam is formed between CO₂ and nanoparticle solution, which suppress the viscous instability.

The results provide nanoparticles are effective to enhance CO₂ fracturing. Also, this experiment suggests an optimized protocol of CO₂ fracturing with nanoparticles in unconventional reservoir stimulate.

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1. Introduction

Over past three decades, the success of shale gas production in U.S. has triggered an increasingly worldwide interest in these unconventional resources. According to U.S. Energy Information Administration (EIA), low permeability gas shale was estimated to hold 800 trillion cubic feet of recoverable natural gas. Economically feasible application of advanced hydraulic fracturing and horizontal drilling technologies in tight gas shale enabled U.S. to surpass Russia to become the top producer of natural gas since 2009. Other countries, such as China, Poland, also possess abundant amount of unconventional reservoir resources. EIA estimates China has 1115 Tcf of risked technically recoverable shale gas resources, making it an important resource for China's future energy demand.

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Most unconventional reservoirs are developed by hydraulic fracturing treatments (Morales et al., 2011; Cheng, 2012; King and King, 2010; King, 2012). In unconventional reservoir fracturing, various fracturing fluids have been applied, such as foam-based fluids, water-based fluids and waterless fluids (Fisher and Warpinski, 2012; Carl Montgomery, 2013). At present, slick water is the mainstay fracturing fluid in unconventional reservoir stimulate. Slick water is fresh water treated with up to 5% potassium chloride by volume. Water fracturing lacks gel particles, therefore it leaves no residues or filter cakes behind, and produces less damage to fracture conductivity compared to massive hydraulic fracturing with gelled fluids (Terracina et al., 2001). However, there are many disadvantages associated with hydraulic fracturing operations. Firstly, huge volume of water is required in hydraulic fracturing which is difficult to achieve in water deficient areas. Secondly, formation damage would occur since a large quantity of aqueous fluid is injected into reservoirs during hydraulic fracturing. Another deficiency is that slick water cannot permeate into the nano-

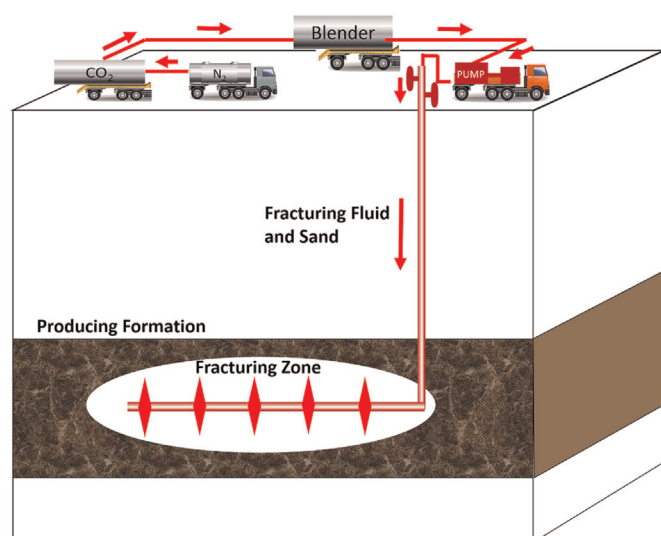
Fig. 1. CO₂ fracturing process.

Table 1
The permeability of shale.

	Eagle ford shale (Gong, 2013)	Bakken shale (Cho et al., 2013)	Barnett shale (Loucks et al., 2009)*
Fracture permeability (D)	0.04–0.2	3–8	0.1–18
Matrix permeability (mD)	0.02–0.8 × 10 ^{−4}	1.31–7.24 × 10 ^{−4}	0.02–3.2

* Calculated based on the data from reference.

Table 2
The character of core.

	Diameter (cm)	Length (cm)	Porosity (%)	Permeability (mD)
Boise sandstone	7.2	30	28.8	900

Table 3
Relevant fluid properties: viscosity μ , density ρ , and interfacial tension with respect to brine σ .

	Brine	5% Nano	CO ₂
μ (cP)	1.1	1.2	0.08
ρ (Kg/m ³)	1010	1040	792
σ (mN/m)	N/A	N/A	24

pores and micro-pores for its high viscosity, which would limit the stimulated reservoir volume (SRV).

Therefore, a proposed fracturing fluid in hydraulic fracturing of unconventional reservoirs is liquid CO₂ (Gupta and Bobier, 1998; Yost et al., 1993). Fig. 1 shows the process of CO₂ fracturing. Liquid CO₂ is more active on extending micro-fractures and connected them during fracturing in shale than water and slick water (Fang et al., 2014). Among the benefits of using liquid CO₂ in hydraulic fracturing is less flow back water that needs to be treated or permanently disposed of. In addition, shale formations are known to preferentially adsorb CO₂ over CH₄ as noted by several researchers (Jessen et al., 2008; Nuttal et al., 2005; Kang et al., 2011; Schepers et al., 2009; Heller and Zoback 2013; Ismail et al., Zoback). Kalantari-Dahaghi (2010) had reported the study of CO₂-EGR and concluded that the process is feasible since CO₂

molecules have greater sorption affinity compared to methane molecules. Thus, another promising application for CO₂ is CO₂ injection for enhanced gas recovery (Eshkalak et al., 2014). Additionally several simulation studies had optimized CO₂ injection process for EOR (Sondergeld et al., 2010; Yu et al., 2014).

In CO₂ injecting to the formation, the less dense and less viscous of CO₂ will cause the poor sweep efficiency by viscous fingering and gravity over-ride (Bae and Irani, 1993; Rossen, 1996; Wagner and Weisrock, 1986). In CO₂ fracturing, as same as CO₂ storage, the viscosity fingers reduce the fracturing fluids drainage area. Recently, there are many researches about using nanoparticles emulsions to enhanced oil recovery (Kotsmar, 2010; Zhang et al., 2009, 2010) And nanoparticle will form a film between CO₂ and brine, which uniform the displacement front to prevent the fingering (Grigg and Schechter, 1997; Aminzadeh et al., 2012a, b; Aminzadeh et al., 2013; Binks, 2002, 2007; Binks et al., 2008; DiCarlo et al., 2011) In this paper, we conducted an experiment to demonstrate the effect of nanoparticles on sweep efficiency and proposed a protocol of CO₂ fracturing.

2. Methodology

In this experiment, Boise sandstone was used instead of shale stone to evaluate the nanoparticle's effect on CO₂ fracturing volume. Shale could be seen as dual-porosity media, including matrix pore system and fracture system. In the flow model, for ultra-low permeability of matrix, it assumes that gas only flowing in fracture system (Apaydin et al., 2012; Hudson et al., 2012; Josh et al., 2012). The permeability and pore size of Boise sandstone are as same as micro fracture and macro pores in shale, potentially having the same conduction (Table 1). Hence, sandstone could be the appropriate media to observe the CO₂ distribution during the injection.

The core is a cylinder that is 7.2 cm in diameter and 30 cm in length. The permeability of core is 900 mD, and porosity is 28.8% (Table 2). In order to avoid CO₂ corrosion, the rock sample was wrapped in a heat-shrinkable Teflon tube, 4 layers of aluminum foil, another layer of Teflon heat-shrinkable tube, and an AFLAS rubber sleeve before it was placed into an aluminum core holder. The Teflon layers provide a barrier to water, while the aluminum foil prevents CO₂ diffusion to the AFLAS sleeve.

The wetting phase as formation water is brine with 2 wt% NaBr. In the accumulator, the brine was pre-equilibrated with CO₂ by injecting 100 ml of CO₂ per liter of brine, into the brine accumulator over 5 h and letting the wetting fluid to equilibrate with CO₂ for 48 h before usage.

In this experiment, CO₂ would be kept in liquid phase at 8.3MPa and 25 °C (room temperature). And CO₂ was saturated with brine by injecting brine (20 ml of brine per liter of CO₂) into the CO₂ accumulator over 5 h and allowing CO₂ and brine to equilibrate for 48 h before usage. The fluids properties are showed in Table 3.

The nanoparticles used in this experiment are made by silicon and coated with polyethylene glycol (PEG), which prevents the aggregation and retention of nanoparticles. The diameter of nanoparticle could ranges from 5 nm to 20 nm. In this experiment we selected 5 nm nanoparticles as test objects. They are small enough to transport into micro-pores of shale stone, which contributes to generate more fractures in matrix. In experiments, the nanoparticle dispersion was diluted to 5 wt% with NaBr aqueous phase whose total salinity is 2 wt%.

3. Procedure

Fig. 2 shows a schematic of our experiment set-up. In this

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