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Potential and challenges for the new method supercritical CO₂/H₂O mixed fluid huff-n-puff in shale oil EOR

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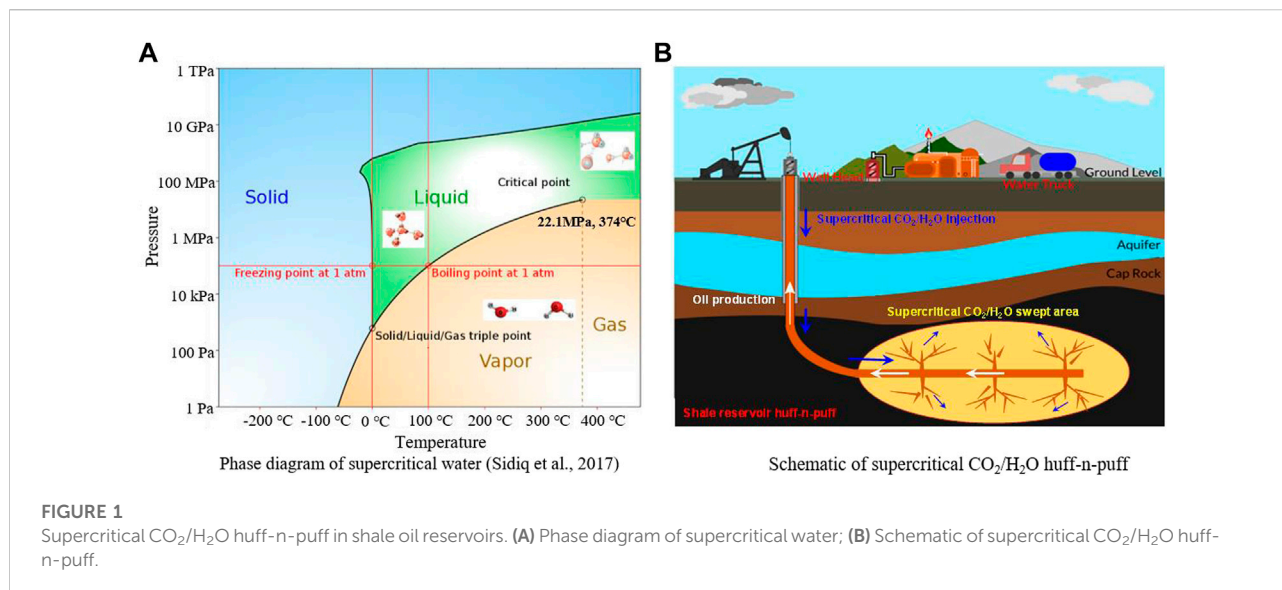
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Introduction

The successful development of shale oil is important to ensure energy security. However, shale oil recovery is typically less than 10%. Supercritical CO₂/H₂O huff-n-puff is a potential EOR strategy for shale oil development, but it is still in the exploratory stage. Supercritical CO₂/H₂O huff-n-puff exerts the capacity of two kinds supercritical fluids, and using the same well as an injection well and a production well, which solves the problem of gas channeling in shale oil reservoir after fracturing by conventional gas drive method. This paper provides a brief overview of the advantages, potential, injection method, summarize the problems and future research directions of the new technology in shale oil development, which is of great important for the shale oil reservoir developments.

Proposal of supercritical CO₂/H₂O huff-n-puff in shale oil reservoirs development

Shale oil is a promising energy source with great potential for development. Shale oil storages in mud shale matrix pores, micro-fractures and thin interlayers of non-source rocks in the occurrence state of free state and dissolved or absorbed state in organic kerogen (Feng et al., 2020; Xu et al., 2022). These characteristics lead to the poor development effect. The movable reserves of shale oil are generally less than 10% under the fracturing conditions of horizontal wells with existing technologies (Hoffman, 2018; Jia et al., 2019). As the low porosity and low permeability, high clay content are key characteristics of shale oil, conventional water injection development is not suitable for shale oil development because of its difficulties in injection, small sweep volume and serious water sensitivity. As the most widely used gas displacement agent, supercritical fluid CO₂ has the potential to develop shale oil efficiently. Compared with CH₄ and N₂, CO₂ is more easily miscible with crude oil, increasing the flow capacity of oil and thus improving oil recovery by 5%–25% (He et al., 2022; Jin et al., 2017; Sheng, 2015).



Supercritical H₂O is a fluid with better performance than supercritical CO₂, with higher dissolving capacity, increasing solvent diffusion coefficient ability and better reactivity, which can improve sweep coefficient and oil washing efficiency in the process of oil displacement (Walther and Woodland, 1993; Schaefer and McGrail, 2004; Li et al., 2020). Meanwhile, the better dissolving capacity and diffusion performance are beneficial for entering nanopores of organic matter, which has the potential to exploit the adsorption and dissolution oil (Weingärtner and Franck, 2005; Caniaz and Erkey, 2014; Zheng et al., 2020). Therefore, the use of supercritical CO₂/H₂O mixed fluid to improve shale oil recovery is considered to be a promising technology.

Status of supercritical CO₂ huff-n-puff in shale oil reservoirs development

In the past few years, supercritical fluid has been applied in many fields as solvent or reaction medium due to its unique properties. Under supercritical conditions, by controlling the pressure, the reaction environment can be manipulated, which can increase the solubility of supercritical fluid in crude oil, enhance the interphase mass transfer ability, and improve the oil stripping effect and oil displacement efficiency. At present, a large number of experiments and numerical simulation studies have been carried out to improve shale oil recovery by supercritical CO₂ (ScCO₂) huff-n-puff. Gamadi et al., (2013), Gamadi et al., (2014) found in laboratory core-scale experiments that the use of nitrogen or CO₂ huff-n-puff at different injection pressures can increase shale core recovery by approximately 10%–50%. And the effect of enhanced oil

recovery relies on injection pressure, huff-n-puff cycles, core size, soaking time and gas injection rate, etc., (Hoffman, 2012; Song and Yang, 2013; Yu and Sheng, 2015; Yu et al., 2016; Zhang, 2016; Li et al., 2017).

Core-scale macroscopic experiments in laboratories have proved that ScCO₂ huff-n-puff is an effective method to improve shale oil recovery. Field tests of ScCO₂ huff-n-puff EOR were carried out in Bakken formation and Eagle Ford in North America. They all reported this method is effective in some extent. However, the ultimate recovery factor of different types of shale rock samples fluctuates greatly after multiple cycles of huff-n-puff. For most shale oil reservoirs, the minimum miscible pressure (MMP) of CO₂ and crude oil is higher than the formation pressure, thus it could not achieve the miscible state, resulting in unsatisfied displacement effect.

The advantages of supercritical H₂O injection fluid

Compared with ScCO₂, the properties of supercritical H₂O (ScH₂O) are more advantageous. As shown in Figure 1A (Sidiq et al., 2017), the condition of ScH₂O is that the temperature and pressure exceed the critical point (22.1 MPa, 374°C), at which time the gas-liquid interface disappears and most of the hydrogen bonds are distorted or broken (Zhao et al., 2019). Because of the absence of surface tension, ScH₂O can enter micropores and nanopores more easily. And in the vicinity of the critical point, the fluid density increases sharply with the increase of pressure, so that the supercritical fluid has a higher density, which is close to the value of liquid. Water exhibits properties closer to non-polar organic compounds and becomes a good solvent for non-polar substances. It can dissolve many solid or high boiling

substances and form a homogeneous phase with organic substances (Adschiri et al., 2011). This feature creates conditions for supercritical water extraction of crude oil.

The viscosity of supercritical water is only one-tenth of that of normal water. The low viscosity makes ScH_2O and solute molecules have higher mobility, and solute molecules can easily diffuse in ScH_2O (Guo et al., 2015; Zheng et al., 2020). Supercritical fluid can not only be used as reaction medium in chemical reaction, but also directly participate in reaction. Supercritical fluid can change the phase behavior, diffusion rate and solvation effect, homogenize the reaction mixture and increase the diffusion coefficient (Heltai et al., 2002; Yang et al., 2019; Chen et al., 2020), thus controlling the phase separation process and the distribution of products. When ScH_2O is used as the reaction medium, its specific physical and chemical properties will affect the progress of the reaction. For example, at 400°C and 35 MPa, the ion product increases to 7 times that of the normal state, which is conducive to the formation of hydrogen bonds. This property can promote the conversion of organic kerogen in shale reservoir and drive oil out of organic matter (Akiya and Savage, 2002; Fauvel et al., 2003).

Therefore, the high dissolving ability, wide diffusivity and strong reactivity of ScH_2O can improve the sweep coefficient and displacement efficiency, thereby improving the oil recovery. The flow characteristics of ScH_2O in the flow process have an important impact on oil recovery, and the heat transfer process from wellbore to formation is an important factor to be considered in the energy utilization process (Nian et al., 2016; Phuoc et al., 2019; Gao et al., 2021). Because of its higher temperature, better cracking rate and larger heating area, ScH_2O flooding shows better effect than hot water flooding and steam flooding (Sun et al., 2018). And in heavy oil production, the operational parameters for supercritical water injection are slightly influencing the reservoir performance, and the influence is negligible as long as a minimum miscible pressure is achieved (Nie, 2021; Ma et al., 2022). As an organic solvent, ScH_2O provides a homogeneous environment for the recovery and thermal cracking of heavy oil. In addition, the properties of ScH_2O can be adjusted over a wide range of temperatures and pressures, which can not only reduce the viscosity of crude oil as a heat carrier, but also eliminate seepage resistance and carbon loss in thermal recovery as a solvent (Zhao et al., 2018).

Status of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff in enhancing oil recovery

ScCO_2 and ScH_2O have great potential for shale oil development. The characteristics of strong mass transfer rate to crude oil, efficient extraction capacity, wide source, easy access of ScCO_2 , combined with the properties of ScH_2O , supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid huff-n-puff can give full play to their

respective advantages. In the process of supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid injection as shown in Figure 1B, the interaction of ScCO_2 and ScH_2O under supercritical conditions (Deleau et al., 2022), the influence of mixed fluid on pore permeability structure (Liu et al., 2020; Zhou et al., 2020) and mechanical properties (Zhou et al., 2019), the interaction of supercritical $\text{CO}_2/\text{H}_2\text{O}$ -shale and its influence on shale adsorption characteristics (Yang et al., 2022) and so on, all play important roles in enhancing oil recovery.

Injection methods of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff

Supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff can be divided into two methods: one is injecting supercritical fluid from the surface; the other one is to inject liquid $\text{CO}_2/\text{H}_2\text{O}$ from the surface and heated in the bottom of the wellbore to achieve supercritical conditions (Fakher and Imqam, 2020; Gao et al., 2022; Ren et al., 2023). For method one, the percolation heat transfer of supercritical fluid in wellbore and reservoir is very complicated, and it is difficult to maintain the high-temperature and high-pressure (HTHP) environment of ScH_2O during transportation. In addition, the mixed fluid is corrosive to the pipe string, so special attention should be paid to the HTHP resistance and corrosion resistance of pipelines in the process of preparation, storage, transportation and injection, which needs advance technology and high requirements for material. For method two, as *in situ* combustion, electromagnetic heating or microwave heating and other formation heat treatment to improve oil and gas recovery is more and more popular in recent years, these treatment methods can be used to achieve the supercritical temperature of CO_2 and H_2O . Thus, the complex heat transfer problems and stringent pipeline material requirements of supercritical fluid injection from wellbore to the reservoir can be solved. In this way, method two is more feasible.

The research direction of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff for shale oil development

Novel laboratory experiments should be designed to study the feasibility of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff in shale oil recovery. 1) The pore structure and mineral composition before and after the interaction of supercritical $\text{CO}_2/\text{H}_2\text{O}$ can be studied by combining nuclear magnetic resonance technology, mercury injection method and scanning electron microscopy test. 2) The interaction between supercritical $\text{CO}_2/\text{H}_2\text{O}$ and crude oil can be obtained by means of HTHP PVT test. 3) Core scale supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff experiments need to be applied to evaluate the EOR performance and the optimization of operation

parameters. 4) The biggest limitation of the existing experimental equipment is that it cannot meet the HTHP conditions of ScH_2O , and the materials of the experimental equipment should be resistant to HTHP as well as corrosion. Nickel-based alloys have excellent strength and corrosion resistance and can be used for pipeline transportation and ScH_2O storage. Therefore, experimental equipment made of nickel-based alloy can be used for indoor HTHP experiments to study the effects of temperature, pressure and injection parameters on the huff and huff effects of supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid. 5) It is necessary to design microscopic experiments and combined with molecular simulation technology to explore the competitive adsorption, dissolution, diffusion and displacement processes in microscopic pores during the huff-n-huff process of supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid, and quantitatively characterize the mobilization mechanism of crude oil in organic matter and inorganic matter.

Conclusion

In conclusion, the mixed fluid of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-puff can give full play to their respective advantages and have great potential for shale oil development. The main challenges of this approach are summarized in detailed below:

- 1) Whether the supercritical condition of $\text{CO}_2/\text{H}_2\text{O}$ can be achieved under shale reservoir conditions is the focus of research. Some formation heat treatment methods such as electromagnetic heating can be combined to meet the supercritical state of $\text{CO}_2/\text{H}_2\text{O}$.
- 2) The interaction between supercritical $\text{CO}_2/\text{H}_2\text{O}$ and shale reservoir is very important for clarifying the mechanism of supercritical $\text{CO}_2/\text{H}_2\text{O}$ huff-n-huff to improve shale oil recovery. The next step is to quantitatively characterize the effects of the two fluids on rocks and pores by means of various experimental studies.
- 3) Laboratory core scale experiment is an important method to verify the feasibility of supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid huff-n-huff to improve shale oil recovery. Materials should be selected to make experimental apparatus suitable for test conditions.

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- 4) Organic nanopores are developed in shale reservoirs. The next research direction is to determine the quantitative characterization methods of dissolution, displacement, diffusion, competitive adsorption and other processes in microscopic pores in the process of huff-n-huff, and to clarify the influence of supercritical $\text{CO}_2/\text{H}_2\text{O}$ mixed fluid on organic matter.

Author contributions

In this article, LL: performed the experiments and wrote the draft. XZ: Literature review, Investigation. YS: Supervision, Investigation. MC: Writing-Reviewing and Editing; PX: contributed to the interpretation and discussion of results; JZ: Literature review, wrote the draft.

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Conflict of interest

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