

## Accepted Manuscript

Ultrasound-Assisted CO<sub>2</sub> Flooding to Improve Oil Recovery

Hossein Hamidi, Amin Sharifi Haddad, Erfan Mohammadian, Roozbeh Rafati, Amin Azdarpour, Panteha Ghahri, Peter Ombewa, Tobias Neuert, Aaron Zink

PII: S1350-4177(16)30338-8  
DOI: <http://dx.doi.org/10.1016/j.ultsonch.2016.09.026>  
Reference: ULTSON 3383

To appear in: *Ultrasonics Sonochemistry*

Received Date: 30 June 2016  
Revised Date: 26 September 2016  
Accepted Date: 29 September 2016

Please cite this article as: H. Hamidi, A. Sharifi Haddad, E. Mohammadian, R. Rafati, A. Azdarpour, P. Ghahri, P. Ombewa, T. Neuert, A. Zink, Ultrasound-Assisted CO<sub>2</sub> Flooding to Improve Oil Recovery, *Ultrasonics Sonochemistry* (2016), doi: <http://dx.doi.org/10.1016/j.ultsonch.2016.09.026>

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.



## Ultrasound-Assisted CO<sub>2</sub> Flooding to Improve Oil Recovery

Hossein Hamidi<sup>a,\*</sup>, Amin Sharifi Haddad<sup>a</sup>, Erfan Mohammadian<sup>b</sup>, Roozbeh Rafati<sup>a</sup>, Amin Azdarpour<sup>c</sup>, Panteha Ghahri<sup>a</sup>, Peter Ombewa<sup>a</sup>, Tobias Neuert<sup>a</sup>, Aaron Zink<sup>a</sup>

<sup>a</sup>School of Engineering, King's College, University of Aberdeen, Aberdeen AB24 3UE, UK

<sup>b</sup>Faculty of Chemical Engineering, University Technology MARA, 40450 UiTM, Shah Alam, Selangor, Malaysia

<sup>c</sup>Department of Petroleum Engineering, Marvdasht Branch, Islamic Azad University, Shiraz, Iran.

### Abstract

CO<sub>2</sub> flooding process as a common enhanced oil recovery method may suffer from interface instability due to fingering and gravity override, therefore, in this study a method to improve the performance of CO<sub>2</sub> flooding through an integrated ultrasound-CO<sub>2</sub> flooding process is presented. Ultrasonic waves can deliver energy from a generator to oil and affect its properties such as internal energy and viscosity. Thus, a series of CO<sub>2</sub> flooding experiments in the presence of ultrasonic waves were performed for controlled and uncontrolled temperature conditions. Results indicate that oil recovery was improved by using ultrasound-assisted CO<sub>2</sub> flooding compared to conventional CO<sub>2</sub> flooding. However, the changes were more pronounced for uncontrolled temperature conditions of ultrasound-assisted CO<sub>2</sub> flooding. It was found that ultrasonic waves create a more stable interface between displacing and displaced fluids that could be due to the reductions in viscosity, capillary pressure and interfacial tension. In addition, higher CO<sub>2</sub> injection rates, increases the recovery factor in all the experiments which highlights the importance of injection rate as another factor on reduction of the fingering effects and improvement of the sweep efficiency.

*Keywords:* CO<sub>2</sub> Flooding; Ultrasound; High frequency waves; Controlled and uncontrolled temperature; Unconventional EOR.

\*Corresponding author at: School of Engineering, King's College, University of Aberdeen, Aberdeen AB24 3UE, UK. Tel.: +44 1224273960. E-mail address: hossein.hamidi@abdn.ac.uk (H. Hamidi).

## 1. Introduction

As oil production declines after primary and secondary recovery methods, tertiary recovery methods need to be considered to further reduce the residual oil saturation in reservoirs. Carbon dioxide (CO<sub>2</sub>) flooding is one of the enhanced oil recovery (EOR) techniques in which CO<sub>2</sub> is injected into oil reservoirs to increase oil recovery. Because of its availability, relatively low cost and environmental considerations, CO<sub>2</sub> is attracting renewed interest as a flooding medium [1]. CO<sub>2</sub> flooding was proposed for the first time in 1930s, and more laboratory and field studies were conducted between 1950s-70s [2] [3] [4] [5] [6]. Those studies concluded that CO<sub>2</sub> could be efficiently implemented to increase oil recovery, since then, CO<sub>2</sub> flooding has become one of the most commonly used EOR methods [7]. On the other side, one of the major problems associated with any flooding process where a high viscous fluid is displaced by a less viscous one, is unstable displacement front. As a result of such instability, viscous fingering develops at the interface of two fluids and starts to grow as the flooding process continues. Therefore, sharp interface of displacement front diminishes by time and early breakthrough of displacing fluid happens. This results in a poor sweep efficiency and a portion of oil remains untouched in the reservoir. This is one of the main concerns in gas injection processes; such as CO<sub>2</sub> flooding enhanced oil recovery method [8]. In 2010, Bagci [9] studied the effect of CO<sub>2</sub> flooding and injection rate on recovery factor in a reservoir with high viscosity oil. It was found that CO<sub>2</sub> breakthrough occurred shortly after the experiment was started. His analysis showed that it was due to the governance of viscous forces and the trivial effects of mass transfer between CO<sub>2</sub> and oil. In addition, the total oil recovery affected substantially for different injection rates due to the associated unstable displacements. Overall, recovery of oil improved at the elevated injection rate of CO<sub>2</sub>.

According to the experimental studies, in addition to the influence of viscous forces, near miscibility condition reduces interfacial tension and oil viscosity, and in turns enhances the final oil recovery [10, 11]. A series of immiscible CO<sub>2</sub> flooding experiments were carried out on heavy oil samples from Wilmington field in United States in 1988 by Mayer *et al.* [12]. The experiments showed that oil recovery was increased due to the reduction of oil viscosity, and oil-swelling. They concluded the improved performance was attributed to more favourable displacement characteristics in the cores. In 2010, Torabi [13] conducted a series of experimental studies to examine the effect of oil viscosity, injection rates, and permeability on the performance of heavy oil water flooding, immiscible CO<sub>2</sub> flooding, and immiscible CO<sub>2</sub> in water alternating gas (WAG) processes. Torabi's experiments revealed that as the viscosity of oil decreases, oil recovery increases for both WAG, and CO<sub>2</sub> flooding processes. Based on Torabi's studies, less viscous oil showed around 42% higher recovery factor than high viscosity oil for CO<sub>2</sub> flooding process. It was also found that in a sand pack with lower permeability, only 25% of oil was recovered. This shows that changes in oil viscosity and permeability of the porous media affect the stability of CO<sub>2</sub> flood front, and in turn the final recovery factor of immiscible CO<sub>2</sub> flooding process. Similar trend of oil recovery was observed when Nasir and Chong [14] performed a number of experiments to study the effect of different CO<sub>2</sub> injection rates on oil recovery in CO<sub>2</sub> flooding process. They showed that recovery factor increases with increasing injection rate. In 2013, Cao and Gu [15] studied the mechanisms of oil recovery from immiscible CO<sub>2</sub> flooding process in core samples of tight sandstone reservoirs. They showed that oil recovery was improved as they increased injection pressure. Their analysis showed that oil recovery improvement was achieved by the reductions of oil viscosity and interfacial tension, and the increase of CO<sub>2</sub> solubility. In 2016, Bikkina *et al.* [16] conducted a set of laboratory

experiments to investigate the effect of reservoir wettability on the efficiency of CO<sub>2</sub> -EOR process. It was found that oil recovery was significantly higher in oil-wet core samples compared to water-wet ones.

On the other hand, techniques such as electromagnetic heating and ultrasound that use electronic means to generate micro- and ultrasonic-waves have been introduced to decrease oil viscosity and mobilize it [17] [18] [19] [20] [21] [22] [23]. Also it was noted that energy through these sources may help to have an efficient oil-water de-emulsification process [24] [25] [26]. The aim of these methods is to deliver an external energy into oil through waves. The transmitted energy can influence physical and chemical properties of hydrocarbons, however, the increase of temperature is one of the most important effects that causes oil viscosity reduction and eventually oil mobilization. In the current study, we limit our investigation into the use of ultrasonic waves for oil recovery processes and, equipment and experiments are designed accordingly. Ultrasound technique as one of the unconventional enhanced oil recovery (EOR) methods was introduced by Duhon and his co-worker [17, 18]. They revealed that ultrasonic waves had a considerable influence on displacement efficiency and oil recovery in the coreflood experiments. Johnston [27] indicated that reductions in viscosity and surface tension of fluids under influence of ultrasound, enhances percolation of oil which causes increase in oil recovery. The reductions were assumed to be due to heat generation by ultrasound absorption in the medium. In fact, ultrasound reduces interfacial tension (IFT) and reservoir oil viscosity, and increases medium temperature, which in turns enhance the displacement efficiency [28, 26, 29]. In 2005, the effect of ultrasound on interfacial tension (IFT) was studied by Hamida and Babadagli [29] by series of pendant drop experiments. They revealed that oil recovery enhancement under influence of ultrasound could be due to a remarkable change in the

interfacial forces between oil and water. In 2012, Mohammadian *et al.* [30] conducted a study on the mechanisms lead to improvements in recovery of oil with the application of ultrasound waves in a waterflooding process. The results show that ultrasound radiation improved oil recovery by 3%. In their study, noticeable temperature rises were recorded during the ultrasonic-assisted waterflooding experiment. This change could affect the properties of fluids such as interfacial tension (IFT) and viscosity, which could be a source of increased mobility of hydrocarbons. In 2013, Abramov *et al.* [31] developed a new technique to improve oil recovery under the influence of ultrasound radiation from failing wells. They showed that ultrasound can noticeably improve oil recovery by 30-50 % from wells where reservoir rock has high porosity (>15%) and permeability (>20 mD). They concluded the technique is environmentally safe and successful for the purpose of oil recovery improvement. Furthermore, in 2015, Abramov and his colleagues [32] performed a series of field tests to develop a sonochemical enhanced oil recovery technique for the treatment of horizontal wells. Their results revealed a significant enhancement in oil production from all wells undergone sonochemical treatment. Later, in 2016, Abramov *et al.* [33] suggested an ultrasound assisted method for treatment of perforation zones in horizontal oil wells (reduction of water cut). A reduction in water cut (20 %) and an increase in oil production up to 91% was reported after the treatment. In recent years, Hamidi and his co-workers [28, 26, 34, 35] performed several experiments to study oil recovery mechanisms under influence of ultrasound in porous media. They found that heat generation, emulsification, cavitation, and the reduction of oil viscosity are the most important mechanisms that improve oil recovery factor under the influence of ultrasound. In their study [28], they indicated that all liquids viscosities were decreased as the result of ultrasonic stimulation and it was more pronounced in uncontrolled temperature condition compared to controlled temperature condition. Furthermore, Hamidi *et*

*al.*'s study on residual oil mobilization in porous media through ultrasonic wave application, suggested a mechanism where alteration of capillary forces is crucial and it is directly proportional to the applied ultrasound power and frequency [34]. In other studies by Hamidi *et al.* [26], they developed a technique to investigate the effect of ultrasound radiation at the interface of oil and brine in porous media using a microscope. Diffusion of phases and generation of emulsion were disclosed in the period that ultrasound was applied. They concluded that emulsification could be another mechanism that improves oil recovery process through short pulses of ultrasound application. Recently, they tested ultrasound assisted surfactant flooding process. Based on phase behaviour analysis of surfactant-brine-oil (SBO), they found that interfacial tension remains low and there is a decrease in surfactant consumption in surfactant flooding processes [35]. Overall the complexity of such process hindered theoretical modelling of ultrasonic enhanced oil recovery technique. For example, Mohsin and Meribout [36] developed a model which involves heat and acoustic modules to predict the performance of ultrasound on oil recovery in a single phase (oil) flow process and they tested their model with their experimental data. They also found that there was a pressure increase due to ultrasonic waves which led to improved oil recovery.

In all previous studies ultrasound was applied to the systems where liquid-liquid or liquid-solid interface were the focus of investigation. However, gas flooding processes could also be potentially good candidates for ultrasound applications. Based on the aforementioned proven mechanisms of ultrasound, combining ultrasound application with CO<sub>2</sub> flooding (Ultrasound-Assisted CO<sub>2</sub> Flooding) in order to improve oil recovery could be of benefit. Therefore, the aim of this study is to identify changes in the oil recovery using ultrasound-assisted CO<sub>2</sub> flooding with different CO<sub>2</sub> injection rates and temperature conditions (controlled and uncontrolled). The

purpose of controlled and uncontrolled temperature conditions was to analyse the influence of ultrasound on the improvement of oil recovery in CO<sub>2</sub> flooding process through heating effects in porous medium. In all the experiments, oil recovery was monitored and the performance of CO<sub>2</sub> flooding, and ultrasound-assisted CO<sub>2</sub> flooding processes were compared.

## 2. Experimental Setup and Procedure

### 2.1 Equipment

In this study, high-frequency waves were produced by a generator (Genesis™ XG-500-6) and delivered to a water bath by means of an immersible transducer (W: 15 cm × L: 35 cm × H: 5 cm). The generator emits ultrasonic waves at a frequency of 40 kHz and a power of 500 W. The ultrasonic bath (W: 21 cm × L: 50 cm × H: 30 cm) was fabricated to provide a suitable medium for ultrasound radiation. The bath efficiency was calculated as 35.4% by the calorimetric technique [37], this indicates the rate of power dissipated in bulk solution is 176.9 W. Ultrasonic bath properties are shown in Table 1. A sandpack was used to represent the porous media and it was immersed into the water bath by a sandpack holder. A syringe pump (KD 100 Scientific) was used for injection of liquid into the sandpack. There is a CO<sub>2</sub> cylinder as a source of gas in the experiments. A chiller (Julabo F25-HL) was used in temperature-controlled experiments to maintain the bath temperature constant. The experimental setup is shown in Fig. 1.

### 2.2 Materials

In this study, paraffin oil was used as oleic phase and its properties are shown in Table 2. The paraffin oil viscosity was measured by Anton Paar™ AMVn Automated Micro Viscometer at two different temperatures (25 °C and 40°C). Carbon dioxide (CO<sub>2</sub>) was chosen as the injection gas in all the experiments. Cylindrical sandpack with the diameter of 5 cm and length of 20 cm



was packed with sands of 60 to 80  $\mu\text{m}$  in diameter. It has a pore volume of 109.95  $\text{cm}^3$ , or simply the porosity of 28%.

### 2.3 Procedure

To run each experiment, first the sandpack was immersed in the water bath to provide a suitable surrounding for ultrasound radiation. Brine (3% NaCl) was injected into the sandpack until it is fully saturated. Then, paraffin oil was injected into the sandpack at a rate of 4.17  $\text{cc}/\text{min}$ . Once the core was saturated with paraffin, water saturation ( $S_w$ ) and oil saturation ( $S_o$ ) was calculated. Then, the sandpack underwent a waterflooding process until no more oil was produced and critical oil saturation was achieved ( $S_{oc}$ ). This was the starting point of the experiments as in this research  $\text{CO}_2$  flooding and ultrasound-assisted- $\text{CO}_2$  flooding were considered as tertiary methods of recovery (EOR) and it was assumed that no more oil could be produced by secondary recovery methods (waterflooding process). However, the authors were aware of different strategies that might be implemented in field operations (EOR from early stages of production). Initial oil and water saturations of sandpack are 0.9 and 0.1 respectively. Waterflooding process was performed on the sandpack, which gave an oil recovery factor of 27%. Residual oil saturation at the beginning of the  $\text{CO}_2$  flooding process was 0.65 with water saturation of 0.35. In tertiary recovery process,  $\text{CO}_2$  was injected at different injection rates (2, 3.5, 5 and 10  $\text{cc}/\text{min}$ ) and the amount of produced oil was measured that is discussed in details in the next section. In ultrasound-assisted  $\text{CO}_2$  flooding experiments, the ultrasonic waves were applied during injection of  $\text{CO}_2$  into the sandpack. Tests were conducted under two different temperature conditions: (i) controlled temperature condition; where temperature inside the ultrasonic bath was maintained constant (about 25  $^\circ\text{C}$ ) by using a chiller, and (ii) uncontrolled temperature condition where there was no control on temperature of the system.

### 3. Results and Discussion

#### 3.1 CO<sub>2</sub> flooding without using ultrasound

The first series of experiments were performed as a benchmark to study the effect of CO<sub>2</sub> flooding on oil recovery improvement without using ultrasound at different CO<sub>2</sub> injection rates. The obtained results were compared to the second series of experiments in which ultrasound was applied. Results of the first series of experiments are presented in Figure 2. It can be seen that the highest oil recovery was achieved for the CO<sub>2</sub> injection rate of 10 cc/min, and the lowest oil recovery for the CO<sub>2</sub> injection rate of 2 cc/min. This is because of the fact that higher CO<sub>2</sub> injection rates can stabilize the interface of CO<sub>2</sub> and oil bank. Higher injection rate of CO<sub>2</sub> provides larger viscous force to push oil and water in the sandpack. Therefore, it creates a larger oil bank, where oil saturation that is left behind interface is low, and as a result the stable flood front reduces the likelihood of viscous fingering and therefore increases the sweep efficiency and oil recovery [14]. Lower injection rates can promote the occurrence of gravity segregation which is a result of large density difference between the less dense CO<sub>2</sub> gas and, significantly more dense oil. In the other words, increasing the CO<sub>2</sub> injection rate can reduce the tendency of gravity segregation of phases, as a larger oil bank is moving toward production wells, in addition, with higher injection rate of CO<sub>2</sub>, it would be easier to overcome the capillary forces in porous media [38, 15]. Therefore, choosing a right injection rate is critical in any CO<sub>2</sub> flooding recovery process to avoid the gravity override problems.

#### 3.2 Ultrasound-assisted CO<sub>2</sub> flooding

In the second series of experiments, the effect of ultrasound-assisted CO<sub>2</sub> flooding on oil recovery improvement in controlled and uncontrolled temperature conditions was investigated. Figures 3 and 4 show oil recovery factors associated with different injection rates in controlled

and uncontrolled temperature conditions respectively. As it is observed, oil recovery was improved in both tested conditions of ultrasound-assisted CO<sub>2</sub> flooding, compared to the cases without using ultrasound (Figure 2). However, these changes were more pronounced for uncontrolled temperature conditions of ultrasound-assisted CO<sub>2</sub> flooding. Furthermore, in these series of experiments, by increasing the CO<sub>2</sub> injection rate, the oil recovery was also improved.

In Figures 5 and 6, we compared oil recovery factor and production rate of three processes investigated in this study for different rates. Comparison of recovery factors and flow rates determines the breakthrough time of CO<sub>2</sub> flood front. For example Figures 5a and 6a show that for the CO<sub>2</sub> injection rate of 10 cc/min, breakthrough of CO<sub>2</sub> flood (end of oil bank production) happens around 12 minutes after the start of the injection process, regardless of type of the process, i.e. breakthrough is unique for ultrasonic-assisted cases and CO<sub>2</sub> flooding without using ultrasonic, and it happens at a later time as CO<sub>2</sub> injection rate decreases.

It can be concluded that higher production rate of uncontrolled ultrasound-assisted CO<sub>2</sub> flooding compared to two other cases is associated with a more stable flood front. This shows that having same breakthrough time for all processes with same CO<sub>2</sub> injection rate, the higher recovery factor means lower residual oil saturation, which means more stable and efficient immiscible displacement process. Therefore, lower residual oil saturation or in the other words, higher sweep efficiency, demonstrates the effectiveness of ultrasonic wave's applications in oil recovery processes. Same trend was observed for other injection rates as can be seen in Figures 5b, 6b; 5c, 6c; and 5d, 6d.

The increase in oil recovery under ultrasound-assisted CO<sub>2</sub> flooding could be attributed to the reduction of oil viscosity. This finding is consistent with that of Torabi's research [13] which revealed that in CO<sub>2</sub> flooding experiments, as the viscosity of oil decreases, oil recovery

increases. Fluid viscosities are critical factors affecting sweep efficiency of any enhanced oil recovery project as they determine the importance of viscous forces in porous media and stability of the moving front. Lower viscosity contrast between displacing and displaced fluids leads to a more stable flood front in immiscible oil recovery processes. This is the main reason for the improvement observed in oil recovery by ultrasound-assisted CO<sub>2</sub> flooding in the current study. In the temperature controlled experiments, the reduction in viscosity of oil could be due to the bubbles implosion produced by pressure impulse as a result of the ultrasound radiation (known as cavitation effect). Cavitation happens in a liquid under the influence of ultrasound radiation when its pressure falls below the vapour pressure, thus generating a bubble or cavity [39]. Injected CO<sub>2</sub> may enter these cavitation zones inside the oil at the interface of oil and CO<sub>2</sub>, which decreases the viscosity, and consequently improves the mobility of oil, and in turn creates a stable interface. In addition, capillary forces are affected by ultrasound radiation in porous media as ultrasonic waves alter the interface shape between two immiscible fluids. Bubbles that are trapped inside the pores during gas flooding absorb the ultrasonic energy and as a result, start to vibrate. The oscillating energy of bubbles may induce a slip effect which improves the flow of fluids inside the pores. This might be explained by the reduction of meniscus effect at the rock-fluids interface and therefore there would be a reduced capillary pressure effect [40, 41]. Fluid slip on solid walls can be described by molecular theories. Fluid displays slip flow when it is in nano-or micro-channels where in these types of channels mean free path of molecules, is comparable to the width of pores [42]. This phenomenon can be induced by ultrasonic waves through the cavitation in oil phase, and it is strengthened inside the pores by penetration of displacing gas molecules, CO<sub>2</sub> in this study, into them. Therefore, a combined effect of viscosity reduction due to ultrasonic waves, CO<sub>2</sub> filled cavities created by ultrasound waves, and capillary

pressure reduction in porous media, might improve the performance of ultrasound-assisted CO<sub>2</sub> flooding process.

Oil recoveries for different CO<sub>2</sub> injection rates shown in Figure 4 are related to the last series of experiments which is ultrasound-assisted CO<sub>2</sub> flooding process in uncontrolled temperature condition. Oil recovery using ultrasound-assisted CO<sub>2</sub> flooding in uncontrolled temperature condition, is improved compared to both temperature controlled CO<sub>2</sub> flooding, and CO<sub>2</sub> flooding without using ultrasound cases for all injection rates. For instance, the ultimate oil recovery using ultrasound-assisted CO<sub>2</sub> flooding in uncontrolled temperature condition with the CO<sub>2</sub> injection rate of 10 cc/min, is 40.9%. It shows ultimate oil recovery of the CO<sub>2</sub> injection rate of 10 cc/min is increased by 7.1 % compared to the case using ultrasound under controlled temperature condition. The only difference between two series of the ultrasound-assisted CO<sub>2</sub> injection is their temperature conditions; one is at constant temperature condition, and the other one at uncontrolled temperature condition, and the temperature profile of the ultrasonic bath through the entire process is shown in Figure 7. Therefore, based on the results, one can conclude that temperature increase at uncontrolled temperature condition, results in a corresponding increase in oil recovery. An increase in the reservoir temperature, results in a reduced interfacial tension leading to improved CO<sub>2</sub> displacement efficiency. As a result of this temperature increase in porous media, oil recovery by CO<sub>2</sub> displacement is improved [30]. These changes which result in a more stable displacement front, through reducing viscosity which in turn diminishes fingering effects and therefore an improved mixing, cause a change in viscous force distribution. Figure 8 shows pressure drop for three cases of experiment with injection flowrates of 10, 5, 3.5, and 2 cc/min. Relatively higher pressure drops are observed for ultrasonic assisted CO<sub>2</sub> flooding cases, furthermore, ultrasonic assisted CO<sub>2</sub> flooding experiments show a

higher pressure drop for uncontrolled temperature case compared to controlled case. It can be concluded that the more stable displacement front, i.e. lower fingering effects, the higher pressure drop to push oil bank ahead of CO<sub>2</sub>. Similar observation reported by Hassan *et al.*'s [43] on the decrease of interfacial tension with temperature at constant pressure condition. They reported the rate of decrease in interfacial tension is more noticeable at higher temperatures. On the other hand, an increase in temperature results in a reduction of oil viscosity leading to an additional improvement of CO<sub>2</sub> displacement efficiency. Thermal energy sources in ultrasound-assisted CO<sub>2</sub> flooding are: (1) the cavitation phenomena by which an enormous amount of thermal energy is released during the collapse of bubbles (known as absorption effect), (2) boundary friction is another phenomena in pore scale, which results in an escalation in temperature of liquid at the solid-fluids interface . Difference in vibration velocity of fluids and rock results in an energy transformation (sound wave to heat) at rock-fluids interface. This happens at both solid-fluids, and suspending particles-fluids interface, and (3) the ultrasound energy dissipation in the porous material [28, 44]. Generally higher wave frequencies correspond to stronger absorption effects and greater boundary frictions. In addition, higher recovery at uncontrolled temperature condition means higher gas saturation in the core which can be concluded that displacement process has a more stable front. Figure 9 summarizes the ultimate oil recovery in all the experiments, using different rates of CO<sub>2</sub> injection.

Further studies such as the use of high resolution microscopes might help us to understand the discussed mechanisms of ultrasonic wave's behaviour in enhanced oil recovery methods. Also it would be interesting to develop a model of ultrasonic waves' effects on CO<sub>2</sub> flooding process to predict the results obtained from our experiments, which can be used to explore and expand this method into the field applications.

#### 4.0 Conclusions

In this study the effect of ultrasonic waves on oil recovery with CO<sub>2</sub> flooding was investigated. Ultrasonic wave's behaviour in a coreflood test showed promising results due to involved complex mechanisms in the molecular and pore scale level. The followings could be concluded based on the experiments conducted in this research:

1. In ultrasound-assisted CO<sub>2</sub> flooding under controlled/uncontrolled temperature condition, oil recovery was improved compared to CO<sub>2</sub> flooding without using ultrasound. This increase could be attributed to mechanisms such as reductions in viscosity, capillary pressure, and interfacial tension.
2. Ultrasonic waves create a stable interface between displacing and displaced fluids, which means reduction of the fingering effects and improvement of the sweep efficiency.
3. More oil was recovered in uncontrolled temperature experiments compared to the controlled temperature experiments.
4. Higher injection rates improved the sweep efficiency through lowering the chance of CO<sub>2</sub> gravity override effects. This means in any CO<sub>2</sub> flooding design, a minimum injection rate needs to be determined to decrease the gravity override issues. The highest recovery was achieved in ultrasound-assisted CO<sub>2</sub> flooding under uncontrolled temperature condition with the highest injection rate of 10 cc/min, resulting in an ultimate oil recovery of around 40.9%.

#### Acknowledgement

The authors would like to gratefully acknowledge and appreciate the School of Engineering, University of Aberdeen, Aberdeen, Scotland, United Kingdom, for the provision of the laboratory facilities necessary for completing this work.

## REFERENCES

- [1] Gaspar, A. T. F. S., Suslick, S. B., Ferreira, D. F. and Lima, G. A. C. , “Economic Evaluation of Oil Production Project with EOR: CO<sub>2</sub>Sequestration in Depleted Oil Field,” *SPE Latin American and Caribbean Petroleum Engineering Conference*, pp. 20-23, 2005.
- [2] L. P. Whorton, E. R. Brownscombe and A. B. Dyes, “Method for Producing Oil by Means of Carbon Dioxide”. USA Patent 2,623,596, 30 December 1952.
- [3] Beeson, S. M., and Orloff, G. D. , “Laboratory Investigations of the Water-Driven Carbon Dioxide Process for Oil Recovery,” *Trans., AIME* , vol. 216, pp. 388-391, 1959.
- [4] Holm, L. W. A , “Comparison of Propane and CO<sub>2</sub> Solvent Flooding Processes,” *AICHE J.*, vol. 7, no. 2, pp. 179-184, 1961.
- [5] Holm, L. W., “CO<sub>2</sub> Slug and Carbonated Water Oil Recovery Processes,” *Producers Monthly*, pp. 6-8, 26-28, September 1963.
- [6] Holm, L. W., and O’Brien, L. J. , “Carbon Dioxide Test at the Mead-Strawn Field,” *J. Pet. Tech.* , pp. 431-442, 1971.
- [7] Farouq Ali, S. M. . , “Non-thermal heavy oil recovery methods,” in *SPE 5893, SPE Rocky Mountain Regional Meeting*, Casper, Wyoming, 1976.
- [8] Orr, F., Heller, J., and Taber, J. , “Carbon dioxide flooding for enhanced oil recovery: Promise and problems,” *Journal of the American Oil Chemists Society*, vol. 59, no. 10, pp. 810A-817A, 1982.
- [9] Bagci, A. S. , “Immiscible CO<sub>2</sub> Flooding through Horizontal Wells,” *Taylor & Francis Group, LLC, Energy Sources, Part A*, vol. 29, p. 85–95, 2007.
- [10] Spivak, A., and Chima, C. M., , “Mechanisms of immiscible CO<sub>2</sub> injection in heavy oil reservoirs, Wilmington Field, CA,” in *SPE 12667, SPE Enhanced Oil Recovery Symposium*, Tulsa, Oklahoma, 1984. April 15–18.
- [11] Ghedan, S. , “Global laboratory experience of CO<sub>2</sub>-EOR flooding,” in *SPE 125581, SPE/EAGE Reservoir Characterization and Simulation Conference*, Abu Dhabi, United Arab Emirates, 2009.October 19–21.



- [12] Mayer, E. H., Earlougher, Sr., R. C., Spivak, A., and Costa, A., , “An analysis of heavy oil immiscible CO<sub>2</sub> tertiary core flood data,” in *SPE Paper 14901, 5th Symp. on Enhanced Oil Recovery*, Tulsa, Oklahoma, USA, 1986.
- [13] Torabi F., “Effect of Oil Viscosity, Permeability and Injection Rate on Performance Of Water Flooding, CO<sub>2</sub> Flooding and WAG Process In Recovery Of Heavy Oils,” in *SPE138188-Canadian Unconventional Resources and International Petroleum Conference*, Calgary, Alberta, Canada, 2010.
- [14] Nasir F. M., and Chong Y. Y., , “The Effect Of Different Carbon Dioxide Injection Modes On Oil Recovery,” *International Journal of Engineering & Technology*, vol. 9, no. 10, pp. 54-60, 2009; 09(10):3-6..
- [15] Cao M., and Gu Y., “Oil recovery mechanisms and asphaltene precipitation phenomenon in immiscible and miscible CO<sub>2</sub> flooding processes,” *Fuel*, vol. 109, pp. 157-166, 2013.
- [16] Bikkina P., Wan J., Kim Y., Kmeafsey T. J., and Tokunaga T. K., “Influence of wettability and permeability heterogeneity on miscible CO<sub>2</sub> flooding efficiency,” *Fuel*, vol. 166, pp. 219-226, 2016.
- [17] Duhon R.D., “An investigation of the effect of ultrasonic energy on the flow of fluids in porous media (Ph.D. thesis),” University of Oklahoma, 1964.
- [18] Duhon, R.D. Campbell, J.M. , “The effect of ultrasonic energy on flow through porous media,” in *SPE 1316:Second Annual Eastern Regional Meeting of SPE/AIME*, Charleston, WV, 1965.
- [19] Aarts A. C. T. , Ooms G. , Bil K. J. , and Bot E. T. G. , “Enhancement of liquid flow through a porous medium by ultrasonic irradiation,” *SPE Journal*, vol. 4, no. 04, pp. 321-327, 1999.
- [20] Hascakir B. , Acar C. , and Akin S. , “Experimental and numerical simulation of oil recovery from oil shales by electrical heating,” *Energy & Fuels*, vol. 22, no. 6, pp. 3976-3985, 2008.
- [21] Bjordalen N., and Islam M. R., “The effect of microwave and ultrasonic irradiation on crude oil during production with horizontal well,” *Journal of Petroleum Science and Engineering*, vol. 43, pp. 139-150, 2004.
- [22] Hascakir B. Acar C. and Akin S., “Microwave Assisted Heavy Oil Production: An Experimental Approach,” *Energy & Fuels*, vol. 23, pp. 6033-6039, 2009.
- [23] Abdulrahman M. M. and Meribout M. , “Antenna array design for enhanced oil recovery under oil reservoir constraints with experimental validation,” *Energy*, vol. 66, pp. 868-880, 2014.
- [24] Mohsin M. and Meribout M. , “Oil–water de-emulsification using ultrasonic technology,” *Ultrasonics Sonochemistry*, vol. 22, p. 573579, 2015.
- [25] Kar T. and Hascakir B. , “The role of resins, asphaltenes, and water in water-oil emulsion breaking

- with microwave heating,” *Energy and Fuels*, vol. 29, no. 6, pp. 3684-3690, 2015.
- [26] Hamidi, H. Mohammadian, E. Asadullah, M., Azdarpour, A. and Rafati R. , “Effect of Ultrasound Radiation Duration on Emulsification and Demulsification of Paraffin Oil and Surfactant Solution/Brine Using Hele-Shaw Models,” *Ultrasonics Sonochemistry*, vol. 26, pp. 428-436, 2015.
- [27] Johnston, H.K., “Polymer viscosity control by the use of ultrasonics,” in *Chemical Engineering Progress Symposium Series 67:39-45*, 1971.
- [28] Hamidi, H. Rafati, R. Junin, R. Manan, M. and Busra, N. , “A Technique for Evaluating the Oil/Heavy-Oil Viscosity Changes under Ultrasound in a Simulated Porous Medium,” *Ultrasonics*, vol. 54, no. 2, pp. 655-662, 2013.
- [29] Hamida, T., and Babadagli, T. , “Effects of ultrasonic waves on immiscible and miscible displacement in porous media,” in *SPE 95327, Presented at the SPE Annual Technical Conference and Exhibition*, Dallas, Texas, 2005.
- [30] Mohammadian E., Junin R., and Rahmani O. , “Effects Of Sonication Radiation On Oil Recovery By Ultrasonic Waves Stimulated Water-Flooding,” *Ultrasonics*, vol. 53, p. 607–614, 2013.
- [31] Abramov V. O. ,Mullakaev M. S., Abramova A. V., Esipov I. B., and Mason T. J. , “Ultrasonic technology for enhanced oil recovery from failing oil wells and the equipment for its implementation,” *Ultrasonics Sonochemistry*, vol. 20, pp. 1289-1295, 2013.
- [32] Abramov V. O. , Abramova A. V. , Bayazitov V. M., Altunina L. K., Gerasin A. S., Pashin D. M., and Mason. T. J. , “Sonochemical approaches to enhanced oil recovery,” *Ultrasonics Sonochemistry*, vol. 25, pp. 76-81, 2015.
- [33] Abramov V. O., Abramova A. V., Bayazitov V. M. , Marnosov A. V., Kuleshov S. P. , Gerasin A. S. , “Selective ultrasonic treatment of perforation zones in horizontal oil wells for water cut reduction,” *Applied Acoustics*, vol. 103, pp. 214-220, 2016.
- [34] Hamidi H., Rafati R., JuninR. B. and Manan M. A., “A role of ultrasonic frequency and power on oil mobilization in underground petroleum reservoirs,” *Journal of Petroleum Exploration and Production Technology*, vol. 2, no. 1, pp. 29-36, 2012.
- [35] Hamidi H., Mohammadian E., Rafati R., Azdarpour A., and Ing J., “Effect of Ultrasonic Waves on the Phase Behavior of a Surfactant-Brine-Oil System,” *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 482, p. 27–33, 2015.
- [36] Mohsin M. and Meribout M. , “An extended model for ultrasonic-based enhanced oil recovery with experimental validation,” *Ultrasonics Sonochemistry*, vol. 23, pp. 413-423, 2015.
- [37] Mason T.J., *Sonochemistry*, first ed., New York,USA: Oxford Science Publications, 1999.

- [38] Mohamed I.M., He J., Nasr-El-Din H. A., “Permeability Change during CO<sub>2</sub> Injection in Carbonate Aquifers: Experimental Study,” in *SPE140979- SPE Americas E&P Health, Safety, Security, and Environmental Conference*, Houston, Texas, USA, 2011.
- [39] Wang Z., and NurA. M., “Effects of CO<sub>2</sub> Flooding On Wave Velocities in Rocks with Hydrocarbons,” *SPE Reservoir Engineering*, vol. 4, no. 4, pp. 429-436, 1989.
- [40] Xu, J. and Attinger D., “Acoustic excitation of superharmonic capillary waves on a meniscus in a planar micro geometry,” *Phys. Fluids*, vol. 19, p. 108107, 2007.
- [41] Radke C. j., Kovscek A.R., and Wong H., “A pore-level scenario for the development of mixed wettability in oil reservoirs. Paper,” in *67th Annual Technical Conference and Exhibition of SPE*, Washington D.C., USA, 1992.
- [42] Herskowitz, M., Levitsky, S., and Shreiber, I. , “Attenuation of ultrasound in porous media with dispersed microbubbles,” *Ultrasonics*, vol. 38, pp. 767-769, 2000.
- [43] Hassan M.E., Nielsen R.F., Calhoun J.C., “Effect of Pressure and Temperature on Oil-Water Interfacial Tensions for a Series of Hydrocarbons,” *Journal of Petroleum Technology*, vol. 5, no. 12, pp. 299-306, 1953.
- [44] Xiao G., Du Z., Li G., Shu Z., “High Frequency Vibration Recovery Enhancement Technology in the Heavy Oil Field of China,” in *SPE 86956 – International Thermal Operations and Heavy Oil Symposium and Western Regional Meeting*, Bakersfiled, California, 2004.

**Table 1**

Properties of the ultrasonic bath.

Type of transducer	Generator	Operating frequency (kHz)	Operating power output (W)	Bath size (W×L×H) (cm)	Calorimetric efficiency (%)
Immersible	Genesis™ XG-500-6	40	500	21×50×30	35.4

**Table 2**

Properties of paraffin oil used in the tests.

Type of oil	Dynamic Viscosity <sup>a</sup> @ 25°C (cp)	Dynamic Viscosity <sup>a</sup> @ 40°C (cp)	Density <sup>b</sup> @ 25°C (g/cm <sup>3</sup> )	API	Thermal conductivity (W/m°C)
Paraffin oil	31.73	17.41	0.73	61.28	0.145

<sup>a</sup>Anton PaarAMVn Automated Micro Viscometer.<sup>b</sup>Precisa XT220A Density Measurement Device.

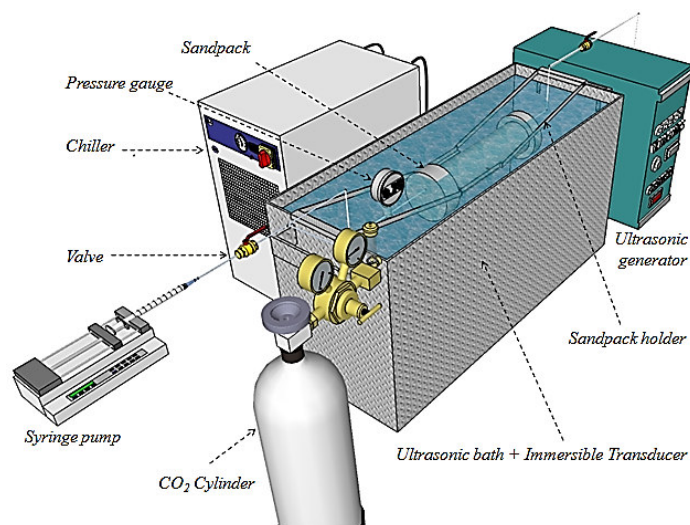


Fig. 1: Schematic diagram of ultrasound assisted  $CO_2$  flooding in controlled temperature condition

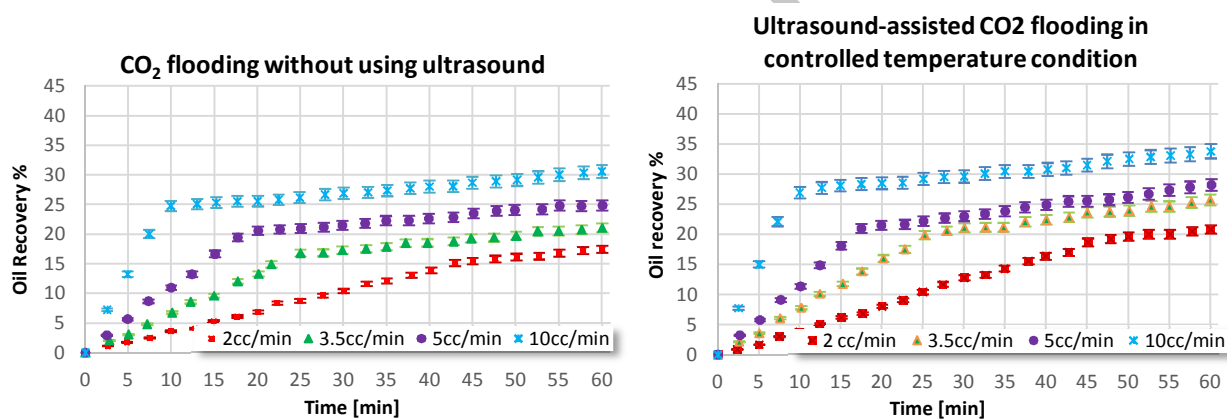


Fig. 2: Effect of  $CO_2$  Injection on oil recovery without using ultrasound

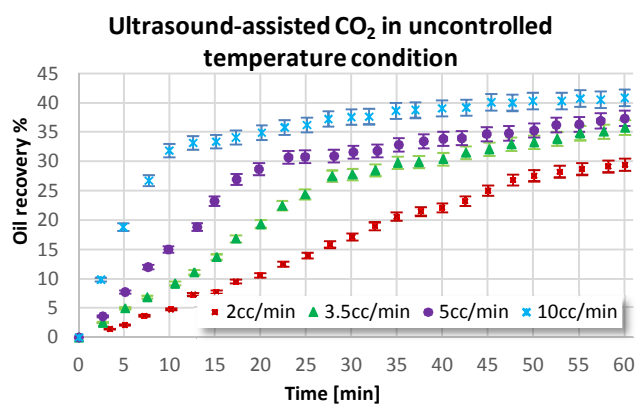


Fig. 4: The effect of ultrasound-assisted  $CO_2$  flooding on oil recovery in uncontrolled temperature condition

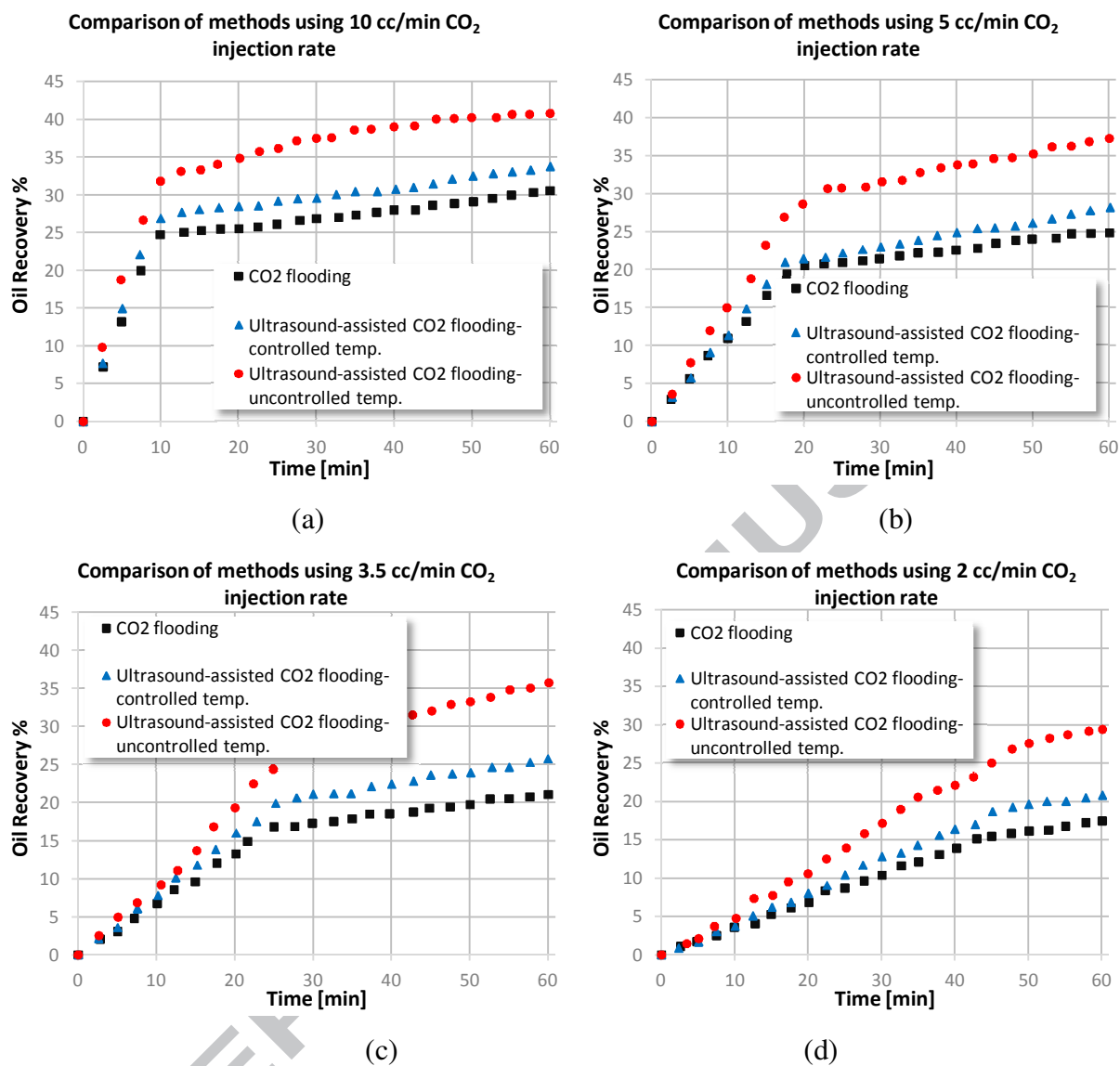


Fig. 5: Effect of CO<sub>2</sub> flooding and ultrasound-assisted CO<sub>2</sub> flooding in controlled/uncontrolled temperature conditions on oil recovery using:(a) 10cc/min,(b) 5cc/min, (c) 3.5cc/min,(d) 2cc/min CO<sub>2</sub> injection rate.

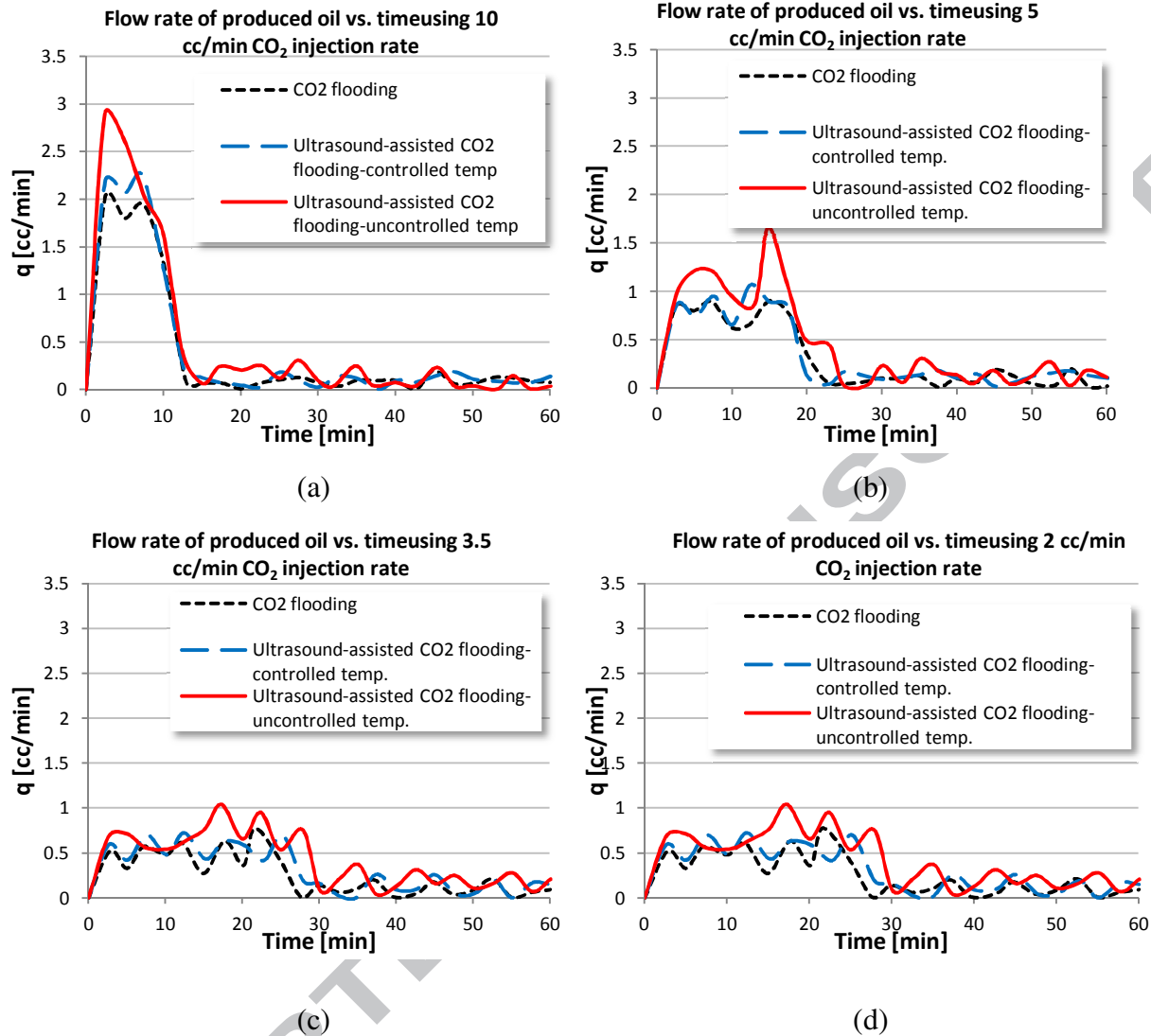


Fig. 6: Flow rate of produced oil for CO<sub>2</sub> flooding and ultrasound-assisted CO<sub>2</sub> flooding in controlled/uncontrolled temperature condition cases using: (a) 10cc/min, (b) 5cc/min, (c) 3.5cc/min, (d) 2cc/min CO<sub>2</sub> injection rate.

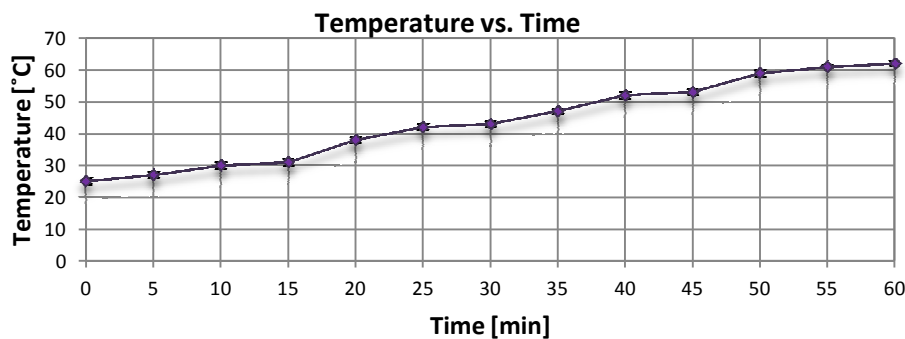


Fig. 7. Ultrasonic bath temperature changes under influence of ultrasonic waves (40 kHz and 500 W)

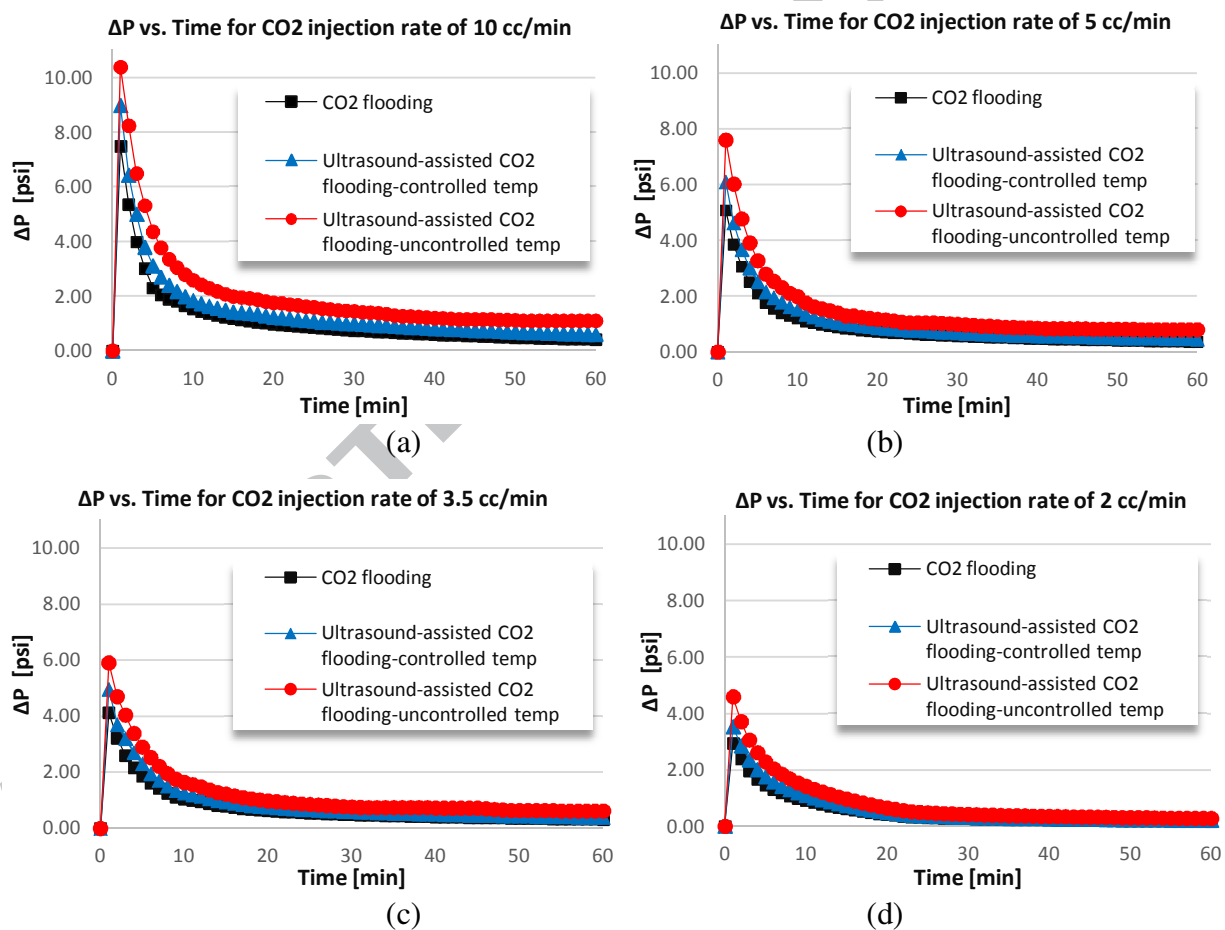


Fig. 8. Pressure drop versus time for CO<sub>2</sub> injection rate of: (a) 10 cc/min, (b) 5 cc/min, (c) 3.5 cc/min, (d) 2 cc/min,



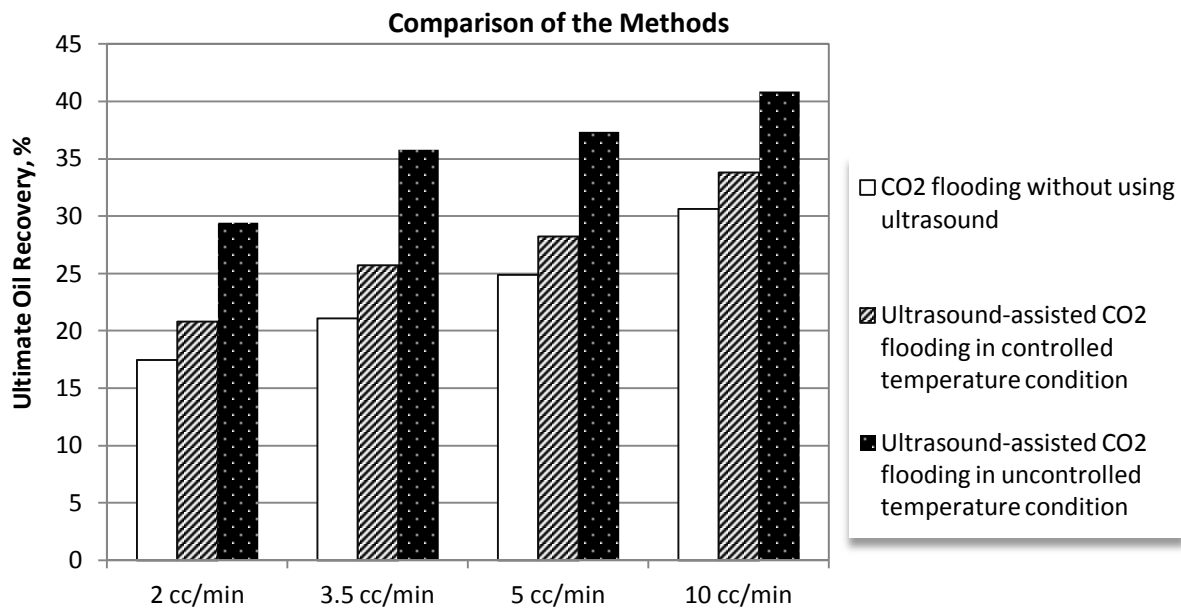


Fig. 9: Results comparison of ultimate oil recovery in all the methods using different CO<sub>2</sub> injection rates

**Highlights:**

- Effect of ultrasound-assisted CO<sub>2</sub> flooding on oil recovery was investigated.
- Experiments were conducted for uncontrolled and controlled temperature conditions.
- Sandpack was put inside the ultrasonic bath.
- More oil was recovered under ultrasound-assisted CO<sub>2</sub> flooding.
- More oil was recovered in uncontrolled temperature experiments.