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# Review article Advances in sour gas injection for enhanced oil recovery-an economical and environmental way for handling excessively produced $H_2S$

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# ABSTRACT

Proper handling of produced sour gas is a big challenge for operating companies due to the serious environmental and technical problems caused by H<sub>2</sub>S. Among various sour gas processing methods, reinjection into reservoirs is a cost-effective and environmentally-friendly technique that not only enhances oil recovery but also allows underground storage of H<sub>2</sub>S. The high safety requirements for working with toxic H<sub>2</sub> S have suppressed extensive experimental studies, hence sour gas injection has not been adequately investigated and implemented in laboratory compared to other enhanced oil recovery (EOR) methods. Existing studies show that sour gas injection with high H<sub>2</sub>S content can bring about higher incremental oil recovery than CO<sub>2</sub> or CH<sub>4</sub> injection due to the highly miscible nature of H<sub>2</sub>S with a significant reduction of minimum miscibility pressure (MMP) and interfacial tension. Mineral dissolution as well as sulfur and asphaltene precipitation might occur as consequences of sour gas induced reactions, thus causing technical challenges such as low injectivity, formation damage, and poor sweep efficiency, but contrary observations were also reported. In addition, the economics of sour gas injection for EOR depends on several critical factors, such as H<sub>2</sub> S content in crude oil, sulfur price, and environmental impact. Overall, sour gas EOR presents far more challenges than other EOR methods, deserving full-fledged and in-depth investigations. This review comprehensively assessed the performance of the sour gas EOR method by integrating the results of laboratory experiments, simulation, field test studies, and economic analysis.

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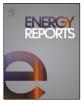
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## 1. Introduction

Natural gas produced from sour oil fields contains a measurable amount of hydrogen sulfide ( $H_2S$ ). The raw gas is defined as sour if the amount of  $H_2S$  in the gas is greater than 100 ppm (Wu et al., 2012). However, this limit varies depending on the jurisdiction and applications. According to the Texas Commission on Environmental Quality, the concentration of  $H_2S$  in raw gas must be greater than 24 ppm to be considered sour.

 $H_2S$  is a highly toxic gas and therefore dangerous for humans. Breathing air with concentrations of  $H_2S$  at 0.1% or 1000 ppm could lead to immediate paralysis of the human respiratory system and death (Draeger Safety, 2013). Interaction of  $H_2S$  with water damages pipeline systems because of corrosion (Wu et al., 2012). In addition, burning the sour gas releases sulfur dioxide, its reaction with water droplets in the atmosphere causes acid rain (Thibeau et al., 2003). Preventing these environmental and technical issues requires proper handling of  $H_2S$  in natural gas, which is a major challenge in the development of sour gas and oil fields.

Carbon dioxide  $(CO_2)$  is also present in sour natural gas.  $CO_2$  exhibits properties similar to those of  $H_2S$  (Wu et al., 2012). Thus, similar methods are applied for removing these gases from the raw gas to meet pipeline and commercial sale requirements (Longworth et al., 1996). This removal process is called "sweetening", and an amine solvent is commonly used to accomplish this. The byproduct of the sweetening process is called acid gas, which contains  $H_2S$  and  $CO_2$  (Wichert and Royan, 1996). Usually, the acid gas is processed in a Claus plant to recover elemental sulfur for sale. However, the Claus process is expensive and ineffective in generating sulfur from  $CO_2$ -rich acid gases (Thibeau et al., 2003). Moreover, the emission of  $CO_2$  into the atmosphere during the Claus process contributes to the greenhouse effect.

Besides the environmental issues, recovering the elemental sulfur has become economically unattractive with the reduction of worldwide sulfur consumption. Sulfur production exhibits an increasing trend due to the development of new sour oil and gas fields. Fig. 1 shows the annual sulfur production in the main sulfur supplier countries in 2020. China Petroleum & Chemical Corporation, Gazprom, Saudi Arabian Oil Co., Abu Dhabi National Oil Company, and Tengizchevroil LLP are the major companies, and produce around 30% of sulfur (Ishtar Company, 2020). Unstable sulfur markets and strict air quality regulations are forcing oil companies to find cost-effective and environmentally friendly methods to handle the acid gas streams.

An alternative approach to generating solid sulfur is to inject compressed gas into appropriate underground formations. Over the last decades, several sour gas injection operations have been successfully implemented in the world, most of them in Canada (Bachu and Gunter, 2005). Advantages of underground acid gas injection over recovering sulfur include zero CO<sub>2</sub> emission, eliminating the expensive Claus process, and insensitivity to acid gas content (Wichert and Royan, 1996). Producing or depleted oil and gas reservoirs and deep aquifers are appropriate zones for acid gas injection (Longworth et al., 1996; Jamaluddin et al., 1998). The proven ability to handle sour or acid gases makes producing or depleted oil and gas reservoirs a good candidate for this purpose. Most of the sour oil and gas fields already have all the required infrastructure for sour gas injection which reduces the cost of the project (Abou-Sayed et al., 2005). The contribution of injected gas to incremental oil recovery by supporting the reservoir pressure and acting as an oil displacement solvent increases the attractiveness of the sour gas injection into oil reservoirs. Nevertheless, the sour gas EOR is relatively new, with numerous uncovered perspectives.

The objective of this paper is to provide information on advances in sour/acid gas injection as an EOR method. This paper is organized in four sections. The first section consists of a brief overview of the thermodynamic properties of sour gas and the influence of  $H_2S$  concentration on the equilibrium between brine/oil/rock systems. The second part presents the effect of  $H_2S$  in the injected gas on the oil phase behavior, minimum miscibility pressure (MMP), and oil recovery based on the results of experimental and simulation studies published. The third section highlights several sour gas injection EOR projects in the world. Comparative economic analyses of various methods available to handle excessively produced  $H_2S$  from sour fields are discussed in the fourth section. The review is concluded with suggestions for future research in this area that should improve our understanding of the sour gas EOR process.

This review does not cover technical issues associated with sour/acid gas injection such as pipeline corrosion and hydrate formation. Mechanical approaches for designing the optimum sour/ acid gas injection schemes, such as surface gas separation, gas injection operations, and water removal, are also beyond the scope of the literature review. Moreover, CO<sub>2</sub> EOR will not be discussed despite the presence of a notable amount of CO<sub>2</sub> in the sour/acid gas.

# 2. Interaction of the sour/acid gas with reservoir fluids and rocks

# 2.1. Physical properties of sour/acid gas

The design of the sour gas EOR process requires a proper understanding of the thermo-physical properties of injected gas compounds. The general properties of  $CO_2$  and hydrocarbon gases have been better established than that for  $H_2S$  due to the high toxicity of the latter. Some of the properties of various gases commonly present in sour gas are listed in Table 1. The critical

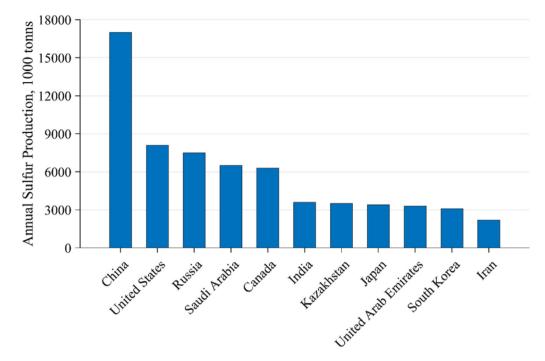


Fig. 1. Annual sulfur production by countries (Fernandez, 2021).

Table 1							
Basic properties	of g	gas p	oresent	in	sour	gas.	

Properties	Gases					
	H <sub>2</sub> S	CO <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub>	$C_2H_6$	
Molar mass, kg/kmol <sup>a</sup>	34.082	44.01	16.043	28.013	30.07	
Critical temperature, °C <sup>a</sup>	100.4	31	-82.6	-146.96	32.2	
Critical pressure, MPa <sup>a</sup>	9	7.38	4.6	3.4	4.87	
Critical density, kg/m <sup>3a</sup>	347	468	163	313.3	207	
Viscosity at 100 °C and 25 MPa <sup>a</sup>	0.091	0.0464	0.0197	0.0257	0.0435	
Density at 100 °C and 25 MPa <sup>a</sup>	661.07	588.45	132.88	201.19	335.5	
Solubility in oil <sup>b</sup>	High	High	Moderate	No	Moderate	
Swells oil <sup>b</sup>	Significant	Significant	Moderate	Low	Significant	
Oil viscosity reduction <sup>b</sup>	Significant	Significant	Yes	No	Yes	
Reduces gas-oil IFT <sup>b</sup>	Significant	Significant	Yes	Yes	Significant	
Forms acid <sup>b</sup>	Yes	Yes	No	No	No	

<sup>a</sup>Values from NIST Chemistry Webbook data (2022).

<sup>b</sup>Information from Burrows et al. (2020).

temperature and pressure of all gases are lower than the reservoir conditions for typical oil fields. Therefore, under reservoir conditions, these gases are either in liquid or supercritical states.

The solubility of gases in oil is linearly correlated to their density, i.e., denser fluid acts as a better solvent for oil. At the same conditions,  $H_2S$  exhibits the highest density. Furthermore, the viscosity of high-pressure  $H_2S$  is approximately two times of that of  $CO_2$  and ethane, and methane is about five times less viscous than  $H_2S$ . Therefore,  $H_2S$  can increase both the density and viscosity of sour gas, which helps prevent gravity override or viscous fingering during gas injection. This leads to better mobility control than other gases, thus can achieve higher sweep efficiency and incremental oil recovery.

# 2.2. Sulfur precipitation

The solubility of elemental sulfur in sour oil is a function of pressure and temperature (Shedid and Zekri, 2002). Sulfur becomes less soluble in the gas or oil phase with decreasing pressure and temperature. When the reservoir conditions reach the sulfur saturation state, the solid sulfur starts to precipitate and further reduction of pressure causes deposition of the sulfur in the formation. The deposition of solid sulfur reduces permeability because of the plugging of porous media and impairs productivity as a consequence (Al-Awadhy et al., 2005).

Shedid and Zekri (2002) conducted a series of core flood tests using a carbonate core sample to investigate the effect of the flow rate, initial sulfur content in oil, and permeability on sulfur deposition. Crude oil samples with a sulfur concentration of 0.78% and 1.76% were used for these experiments. Results showed that the high initial sulfur content in the oil increases the risk of pore plugging, doubling the sulfur content decreased the permeability up to 10% more at the same flow rate. In another work, Shedid and Zekri (2006) highlighted that simultaneous increase in sulfur and asphaltene content damaged the rock permeability. Nonetheless, separate effects of sulfur and asphaltene were not investigated so it is difficult to evaluate the contribution of each component in permeability reduction. Xiao et al. (2010) reported that the adverse effect of sulfur deposition on permeability is significant in low permeability oil reservoirs due to the higher shear rate. On the contrary, Alawi et al. (2015) observed that effect of H<sub>2</sub>S content on water-oil relative permeability was negligible during CO<sub>2</sub> and CO<sub>2</sub> water-alternating-gas flooding, where H<sub>2</sub>S concentration in crude oil was up to 12%. These studies indicate

that sulfur precipitation depends on several factors, such as H<sub>2</sub>S content, crude composition, rock properties and flow conditions.

#### 2.3. Asphaltene precipitation

Gas flooding can also destabilize asphaltene in oils. This destabilization leads to the precipitation of solid asphaltene granules, which plug the porous medium and restrict fluid flow in the reservoir. Non-polar gases such as  $CO_2$ ,  $N_2$  and  $CH_4$  act as strong asphaltene precipitants because of their low polarizability (Yonebayashi et al., 2016). Compared to  $CO_2$ , a very limited number of studies on the effect of  $H_2S$  on asphaltene precipitation have been carried out. Bennion et al. (1999) mentioned that asphaltene precipitates from oil due to mixing with rich acid gas, which has severe effects on permeability. Sayegh et al. (2007) observed that acid gas with 20%  $H_2S$  caused more asphaltene precipitation compared to pure  $CO_2$  by conducting a set of experiments using rising bubble apparatus (RBA).

However, Van Vark et al. (2004) defined acid gas as a good asphaltene solvent that helps to mitigate the risk of asphaltene precipitation, as it decreases the asphaltene onset pressure. In particular, Yonebayashi et al. (2016) conducted a compositional flow simulation to study the impact of the acid gas composition on asphaltene stability, with the expectation that the high polarizability of H<sub>2</sub>S offsets CO<sub>2</sub>-induced asphaltene precipitation. For the base model, crude oil with 15% H<sub>2</sub>S concentration was used. Sensitivity analysis was performed by increasing only the H<sub>2</sub>S concentration from 15% to 76% or the CO<sub>2</sub> concentration from 4% to 6%, as well as increasing the concentration of both gases simultaneously. These analyses showed that increasing the H<sub>2</sub>S concentration narrows the area of the asphaltene precipitation envelope and moves it toward a low-pressure and lowtemperature zone, preventing asphaltene precipitation, while CO<sub>2</sub> has an adverse effect. The influence of CO<sub>2</sub> on asphaltene precipitation was not significant due to its low concentration. Hence, further study is required at higher CO<sub>2</sub> concentrations. Despite of the effectiveness of H<sub>2</sub>S in mitigating asphaltene precipitation proved by simulations, experiments are recommended for verification. As can be seen, the influence of H<sub>2</sub>S on asphaltene precipitation behavior is still questionable and requires further studies.

#### 2.4. Mineral dissolution

When the injected sour/acid gas contacts connate water, acidic compounds partially dissolve in the water, forming an acidic environment and reducing the pH (Xiao et al., 2010). The low pH environment accelerates the dissolution of carbonate minerals and cement in the matrix, which may increase porosity and permeability (Vickerd et al., 2006; Xiao et al., 2009). Such reactions could also free the insoluble fines in the matrix and cement, which may damage the permeability as illustrated in Fig. 2 (Bennion et al., 1999).

Nevertheless, Davison et al. (1999) mentioned that the possibility of dissolution and precipitation due to reinjection of associated acid gas into the sour oil reservoir is low. Based on CT scan images taken during acid gas injection, Vickerd et al. (2006) observed that the porosity and permeability increased due to water desiccation by the injected gas and fines dislodgement but not mineral dissolution. In this study, the water saturation was assumed to be 20%, and the injected gas stream contained 81% H<sub>2</sub>S and 15% CO<sub>2</sub>. By applying new thermal physical and chemical equilibria, Ceragioli and Gianelli (2008) reported that the effect of mineral dissolution on porosity during acid gas injection into carbonate sour oil reservoirs was insignificant because of the limited amount of immobile connate water. In another experimental study, Obeida et al. (2009) observed that acid gas injection (80% CO<sub>2</sub> and 20% H<sub>2</sub>S) for displacing oil had a negligible effect on permeability of dolomite and limestone core samples despite the changing flow rates and 48 h of soaking period. From the above studies, the dissolution of minerals, mostly carbonate, caused by geochemical interaction in the gas/water/rock system during sour/acid gas injection is negligible in oil reservoirs, given very low connate water saturation. It should be aware that the injection of acid gas close to an oil–water contact or aquifer may lead to continuous calcite dissolution and a significant increase in porosity as a result of the continuous flow of water. Another analog would be a simultaneous injection of the acid/sour gas and water or water-alternating-gas process.

# 2.5. Solubility and oil swelling

Both  $H_2S$  and  $CO_2$  are diatomic gases and exhibit high solubility in hydrocarbons (Bennion et al., 1999). At a pressure of 50 MPa and a temperature of 100 °C, the solubility of  $CO_2$  in water is less than 40 m<sup>3</sup>/m<sup>3</sup>, while in the API 40 oil, the solubility of  $CO_2$  increases to 175 m<sup>3</sup>/m<sup>3</sup> (Jamaluddin et al., 1998). The higher solubility of the rich acid gas in the oil has potential benefits on EOR due to the swelling effect and viscosity reduction (Davison et al., 1999). The injected acid gas comes into contact with trapped oil and dissolves into it, thus expanding the oil volume and making it mobile. An adverse effect of oil swelling on permeability can occur if the oil saturation is lower than the irreducible level. In this situation, the trapped oil expands upon contact with the acid gas but cannot reach the irreducible level, so it occupies more pore space and impairs permeability.

Bennion et al. (1999) reported that the injection of a hydrocarbon gas mixture with 50%  $H_2S$  and 50%  $CO_2$  acid gas increased the condensate swelling factor by 20% when the acid gas concentration reached 40%. Luo et al. (2019b) observed that  $H_2S$  exhibited a similar swelling factor to that of  $CO_2$  upon mixing with oil. In their study, the  $H_2S$  swelling factors for volatile and black oil samples were surprisingly almost identical, but the reason was unknown. Besides that, the presence of  $CH_4$  or  $N_2$  as impurities in  $CO_2$  reduced oil swelling, where the effect of  $N_2$  was stronger. Only binary gas mixtures were used in both studies mentioned above; therefore, the influence of sour gas content on the swelling factor should be studied.

# 2.6. Effect of sour/acid gas on oil phase behavior

Understanding the phase behavior of the injected gas and crude oil mixture plays a significant role in designing the sour gas injection process. The pressure–temperature diagram is widely used to examine the phase behavior alteration of the reservoir oil upon interaction with injected gas. Recently, multiple studies have been conducted to investigate the influence of H<sub>2</sub>S on the phase behavior of reservoir oil.

He et al. (2019) analyzed how various gases (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>S, and associated sour gas) affect the phase behavior of crude sour oil, which contains approximately 14.69% H<sub>2</sub>S and 4.12% CO<sub>2</sub>. The molar composition of H<sub>2</sub>S and CO<sub>2</sub> in the associated gas was 0.1742 and 0.0492, respectively. The injection gas and crude oil were mixed in the mole ratio of 0.3. Fig. 3 shows the phase envelope curves for various gas injection cases. Phase behavior analysis demonstrated that injecting H<sub>2</sub>S significantly lowers the pressure at the bubble point and results in the smallest phase envelope of crude oil, while injection of methane has an opposite effect. Small changes were observed after injection of the associated gas and CO<sub>2</sub>. These gases slightly increased the bubble point pressure upon contact with the crude oil. The relationship

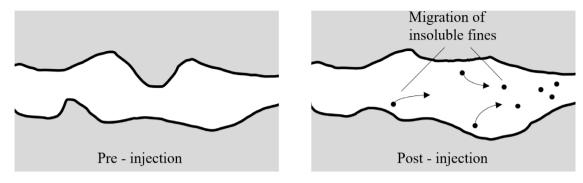
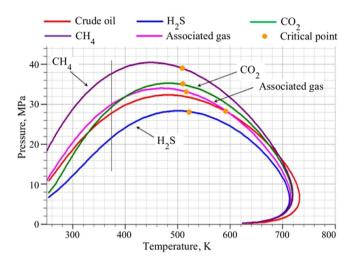


Fig. 2. Illustration of permeability alteration due to dissolution.



**Fig. 3.** Influence of gas composition on the phase diagram (injection gas mole fraction f= 0.3) (He et al., 2019).

between the phase behavior and the mole fraction of the injection gas revealed that crude oil becomes more volatile with the increasing ratio of gas injected. Particularly, a higher ratio of injected associated gas (59% CH<sub>4</sub>, 8.7%  $C_2H_6$  and 8%  $C_3-C_4$ ) increases the composition of intermediate hydrocarbons in the crude oil, leading to higher shrinkage.

Using crude oil with approximately similar acid gas content, Wang et al. (2020) observed that crude oil with H<sub>2</sub>S exhibited the smallest two-phase region and the lowest bubble point pressure, while replacing the H<sub>2</sub>S with CO<sub>2</sub> or CH<sub>4</sub> enlarged the two-phase region and shifted the bubble point pressure curve up. At the same temperature, the bubble point pressure increases in the following trend: H<sub>2</sub>S < CO<sub>2</sub> < CH<sub>4</sub>. Both of these studies showed that H<sub>2</sub>S-rich gas is more favorable for achieving miscibility than CO<sub>2</sub> or CH<sub>4</sub> and allows the prevention of free gas formation due to the lower bubble point pressure. However, in both cases, the influence of the crude oil composition on the phase behavior was not considered.

Luo et al. (2019b) used sour volatile oil, volatile oil and the black oil samples to investigate the effect of the oil composition on the phase behavior, when various gases ( $CO_2$ ,  $H_2S$  and  $CH_4$ ) were injected.  $H_2S$  and  $CO_2$  constituted more than 19% of the mole fraction of the sour volatile oil. Similar to the above studies,  $H_2S$  resulted in the smallest two-phase region and the lowest bubble point pressure, which is followed by  $CO_2$ .  $N_2$  and  $CH_4$  led to a higher bubble point pressure, where the  $N_2$  effect was stronger. It was stated that the volatile oil and the sour oil exhibited approximately similar phase envelopes despite the presence of the acid gas components in the latter. Nonetheless,

the phase envelopes of the black oil were different. For the black oil, all the bubble point curves were in a downward shape, while both samples of volatile oil exhibited expanding ends in the lowtemperature region on the curves when in contact with CH<sub>4</sub> and CO<sub>2</sub>. This difference in the curves revealed that the composition of the crude oil contributes substantially to the alteration of the phase behavior.

## 3. Recovery performance of sour/acid gas injection

#### 3.1. Effect on the minimum miscibility pressure

One of the important parameters for designing a gas injection EOR project is the minimum miscibility pressure (MMP). The MMP is the lowest pressure at which in situ multi-contact miscibility can be achieved between the injected gas and reservoir oil. The primary methods used to determine MMP include (Trivedi et al., 2007):

- Slim tube displacement method
- Rising bubble or falling drop methods
- Mixing cell method
- Compositional simulation of the slim tube test
- Analytical methods
- Empirical correlations

The widely used and generally accepted experimental method is a displacement test on slim tube apparatus, which consists of a small diameter coiled tube packed with sand grains or glass beads (Metcalfe, 1982). Even so, the slim tube results are misleading to some extent because of the lower level of physical dispersion than in the field (Johns et al., 2000). Another experimental method for measuring MMP is to use a rising bubble apparatus (RBA), in which a gas bubble is injected into the bottom of the tube filled with oil and water columns. The shape of the bubble is monitored through a window on the column while the bubble is rising up and dispersing into oil because of mass transfer between the gas bubble and the oil. In most cases, RBA provides reliable and accurate results only for vaporizing drive miscibility mechanism. Therefore, for condensing drive or condensing/vaporizing systems, it is recommended to use the falling drop method (Zhou and Orr, 2013), in which the oil drop falls through the gas phase. Measurement of MMP using RBA or the falling drop method is cheaper, takes less time, and requires less volume of sample compared to the slim tube test (Green and Willhite, 2018). Despite that, difficulties in the visual determination of miscibility and lack of porous media make these methods less accurate than the slim tube method (Dindoruk et al., 2021).

Microfluidic method or slim-tube method on a microfluidic chip (Fig. 4) have been recently developed for MMP determination at reservoir conditions. Nguyen et al. (2015) applied a fluorescence-based microfluidic method to estimate the MMP of a

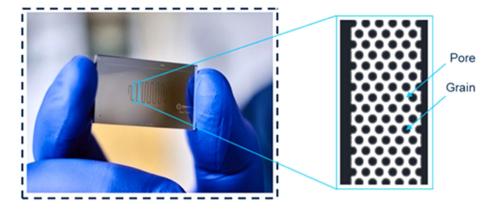


Fig. 4. The microscale slim tube chip used for MMP determination by Ungar et al. (2021), with permission from Elsevier.

 $CO_2$ -oil system. Zou et al. (2019) used a microfluidic chip, which has a single microchannel with several pockets, to study the development of the miscibility for the ethanol-water-hexanol system at ambient conditions. Ungar et al. (2021) demonstrated a novel microfluidic method to measure the MMP at reservoir conditions during oil displacement by hydrocarbon gases and CO<sub>2</sub>. The application of a microfluidic slim tube apparatus significantly reduces the sample volume, which makes the test faster and less costly. Besides that, the micromodel allows one to visualize the flooding process at pore scale, which is impossible in conventional slim tube experiments. The microfluidic method provides fast and cheap opportunities to perform more MMP measurements for early EOR screening. Most importantly, the exceptionally small oil and gas sample volume needed in the microfluidic tests makes MMP measurements with the poisonous sour or acid gases in a regular lab possible.

Empirical correlations and analytical methods are fast compared to the experimental approach. Prediction precision of the empirical correlations for a specific fluid reckons on how close it is to the reservoir fluid properties that were used to develop the correlations (Jarrell et al., 2002). Although analytical methods are based on an accurate EoS, this requires good matching with PVT data (Dindoruk et al., 1997). Similar to the analytical methods, compositional simulation relies on EoS and PVT data but is more computationally expensive and depends on the correct grid size. Non-experimental methods for MMP calculation are cheap and fast but require careful calibrations.

Metcalfe (1982) conducted slim tube experiments to investigate the influence of  $H_2S$  and  $CH_4$  on  $CO_2$  MMP. A sample from the oil field in Texas, which was free of  $H_2S$  and contained approximately 65 mol%  $C_{7+}$ , was used. The set of displacement tests were carried out with mixtures of  $CO_2$ ,  $H_2S$  and methane. The mole ratio of  $CO_2/H_2S$  was assumed as 3:1 and 1:1 in all gas mixtures, while the concentration of  $CH_4$  was varied from 0 mol% to 20 mol%. The  $CO_2$  MMP became higher with the increasing  $CH_4$  concentration.

The experiments showed that MMP decreases with increasing  $H_2S$  content and this effect becomes stronger at higher methane concentrations, but on the percentage-basis, the MMP reduction was almost constant. A 3:1 mole ratio of  $CO_2/H_2S$  reduced the MMP by 18% at each concentration of  $CH_4$ , while a reduction of approximately 30% was reached in the case of a 1:1 mole ratio. In comparison,  $H_2S$  had the same contribution to the reduction of MMP as ethane, and exhibited less efficiency than propane and butane.

Similar to the above study, Lv et al. (2016) investigated the influence of the acid gas composition on the MMP by conducting a slim tube experiment. The acid gas sample containing 78%  $H_2S$  and 22%  $CO_2$  was mixed with the solution gas at various

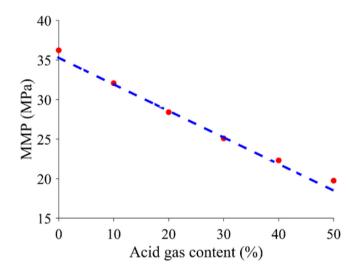


Fig. 5. MMPs with various acid gas compositions (Lv et al., 2016).

proportions prior to injection. The reservoir oil and the solution gas were free from  $CO_2$  and  $H_2S$ . The results of the MMP values are demonstrated in Fig. 5, which shows that the MMP decreases linearly with the increasing amount of acid gas in the solvent. Sayegh et al. (2007) reported a linear relationship between the MMP and  $H_2S$  concentration in a  $CO_2$  mixture, based on the results of experiments using RBA.

Xu et al. (2015) applied EoS-based calculation to determine the MMP for injecting methane, H<sub>2</sub>S, CO<sub>2</sub> and ethane. In contrast to previous studies, the calculation showed that the MMP of H<sub>2</sub>S was higher than the MMP of CO<sub>2</sub> at the same concentration in the solvent. The study revealed that the extraction ability of CO<sub>2</sub> was stronger than that of H<sub>2</sub>S because the molecular weight (MW) of CO<sub>2</sub> was closer to the MW of crude oil. In another similar work, Kaita et al. (2020) performed a comparative study of the MMP for various gases by applying Glaso's (1985) and Yuan et al. (2004) correlations. Both correlations revealed that adding H<sub>2</sub>S to the binary mixture of CO<sub>2</sub> and CH<sub>4</sub> resulted in a slightly higher MMP than the control mixture without H<sub>2</sub>S. This deviation in the empirical results of previous experimental studies could be associated with the inaccuracy of these correlations. Because both correlations were designed for CO<sub>2</sub> injection with impurities such as hydrocarbon gases and nitrogen, they are not applicable for the presence of H<sub>2</sub>S.

He et al. (2019) used the analytical mixing cell method to calculate the MMP of the sour oil after injection of CO<sub>2</sub>, H<sub>2</sub>S, methane, and associated sour gas. The associated sour gas contained 17.42% H<sub>2</sub>S, 4.92% CO<sub>2</sub> and 59.03% CH<sub>4</sub>. Fig. 6 shows that

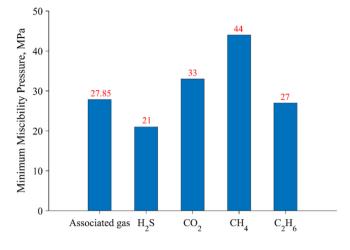
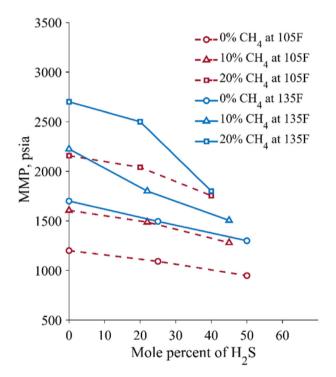


Fig. 6. Mixing cell method results for various gas types (He et al., 2019).



**Fig. 7.** Effect of  $H_2S$  concentration on MMP (1 - Metcalfe (1982); 2 - Sayegh et al. (2007)).

the injection of  $H_2S$  gave the lowest MMP, while the injection of  $CH_4$  resulted in the highest MMP. The miscibility of the associated sour gas and ethane with oil was almost similar. The MMP for the injection of  $CO_2$  was between the associated sour gas and methane. Moreover, it was observed that the miscibility of sour gas with oil becomes weaker with decreasing  $H_2S$  content.

These works showed that the miscibility of  $H_2S$  with oil is greater than  $CO_2$  and the injection of sour gas with  $H_2S$  increases the possibility of achieving miscibility. Besides that, adding  $H_2S$ is beneficial to generate  $CO_2$  miscible displacement because of additional MMP reduction by the  $H_2S$ , which results in higher oil recovery than pure  $CO_2$  injection. The results of several experimental works are summarized in Fig. 7.

# 3.2. Mechanism of sour gas displacement

The achievement of miscibility between the injected gas and in situ oil is a key consideration for gas flooding efficiency. Miscibility is conveniently developed through two different processes, namely the first-contact miscibility (FCM) and multi-contact miscibility (MCM). If the injected gas is fully dissolved into the oil upon the first contact, it is FCM. If the miscibility is dynamically developed in situ through multiple contacts, it is MCM. The MCM process is divided into vaporizing, condensing, and vaporizing/condensing processes. Traditionally, during the vaporizing drive process, the intermediate components of the oil vaporize into the light hydrocarbon-injected gas and enrich it repeatedly through multiple contacts until miscibility is achieved. During the condensing process, the intermediate component of the injected gas condenses into the oil and modifies its composition until miscibility is developed. In the latter case, the injected gas contains intermediate-molecular-weight hydrocarbons. That is why it is expensive but requires less pressure than the vaporizing drive to reach miscibility (Green and Willhite, 2018). However, Zick (1986) reported that the condensing drive mechanism is a combination of vaporizing and condensing gas drive processes, rather than just condensing, which results in better displacement efficiency, although true miscibility may never be fully developed.

Luo et al. (2019a) performed a simulation study using the cell-to-cell method presented by Metcalfe et al. (1973) and Pederson et al. (1986). They investigated multiple contact miscibility mechanisms between reservoir oil and mixed gases with various concentrations of CH<sub>4</sub>, H<sub>2</sub>S, and CO<sub>2</sub>. Additionally, the FCM and MCM pressures for various gases were quantitatively estimated using the pseudoternary diagram and pressure composition diagrams. An oil sample consisting of 15.1% H<sub>2</sub>S and 4.13% CO<sub>2</sub> from the Pre-Caspian carbonate reservoir was selected for this study. The simulation study revealed that the MCM and FCM pressures become lower with the increasing concentration of H<sub>2</sub>S to the pressure reduction was greater than that of CO<sub>2</sub>. On the contrary, CH<sub>4</sub> increased both the MCM and FCM pressures.

Fig. 8 illustrates the MCM and FCM values for different concentrations of  $CO_2$  and  $H_2S$  with mole fractions of  $CH_4$  of 0, 0.25, 0.50 and 0.75. These figures demonstrate that the effect of  $H_2S$ on MCM and FCM was sensitive to the methane concentration in the gas. When the mixed gas contained 0 or 0.25 moles of methane, the MCM was significantly reduced with increasing  $H_2S$  concentration in proportion to the reduction in  $CO_2$ , while the pressure of FCM was kept constant. When the mole fraction of methane reached 0.50 and 0.75, the change in the pressure of the MCM was insignificant and the FCM decreased with the increasing proportion of  $H_2S$ . The reduction in both the MCM and FCM caused by  $H_2S$  was observed to follow a linear trend. It was reported that the miscibility mechanism for the gas produced containing 16%  $H_2S$  and 5.5%  $CO_2$  is a vaporizing process rather than a condensing one based on the ternary diagram.

In another work, He et al. (2019) conducted a series of 1D slim tube test simulations in Eclipse 300 to study the miscible mechanism of crude oil displacement by associated sour gas. A sour crude oil sample from the K oil reservoir in Kazakhstan was selected. The crude oil composition was similar to the oil sample in the study by Luo et al. (2019a). The associated sour gas contained 17.42% H<sub>2</sub>S and 4.92% CO<sub>2</sub>. The gas, transition and oil zones were observed in the gas saturation profile within the slim tube at 0.4 pore volume (PV) injected gas volume and at a pressure that was slightly greater than the MMP. The interfacial tension within the transition zone first decreased until the minimum and then increased. The IFT profile in Fig. 9 shows that the displacement of oil with the associated sour gas is a combined vaporizing/condensing

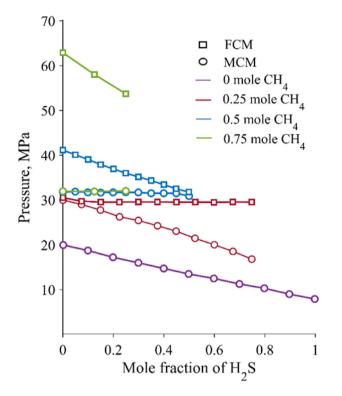


Fig. 8. MCM and FCM for various mole fractions of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  (Luo et al., 2019a).

process, where the vaporizing drive is slightly dominant. In the vaporizing drive mechanism, the vapor was enriched with intermediate components such as  $C_7-C_{20}$  from the crude oil, while in the condensing drive mechanism, light vapor components such as  $C_2-C_6$ ,  $CO_2$  and  $H_2S$  condensed into the crude oil. However, near miscibility occurred in the middle of the transition zone, because zero IFT could not be reached. Further increasing of the pressure narrowed the transition zone, which eventually disappeared at 36 MPa. This indicated that the injected gas completely dissolved in the crude oil at the first contact.

In a later work, Luo et al. (2019b) pointed out that the injected gas composition is responsible for establishing the type of miscibility mechanism, rather than the oil composition. Three oil samples were used in their study, namely sour volatile oil, volatile oil and black oil. A binary mixture of  $CO_2$  with  $H_2S$ , nitrogen, and methane was selected to mix with oil samples. The pseudo pressure diagrams of the mixtures of the oil and gas samples showed that miscibility for the  $CO_2/H_2S$  mixture was developed by a condensing process, while vaporizing was attributed to  $CO_2/N_2$  and  $CO_2/CH_4$ . Fig. 10 shows that increasing the  $H_2S$  content in the  $CO_2$  gas mixture reduces the pressure of the MCM, whereas  $N_2$  and  $CH_4$  lead to an increase in the MCM. However, more studies with three and more impurities in the  $CO_2$  mixture are needed to investigate the influence of the gas type on the miscibility mechanism.

#### 3.3. Laboratory studies on sour/acid gas EOR

In the literature, laboratory studies focusing on the EOR performance of sour/acid gas injection are limited because of the toxic nature of  $H_2S$ . Despite the difficulties in working with sour gas, however, several experimental studies have been carried out. Jacobson (1972) conducted a series of displacement tests to determine the contribution of ethane, propane, butane, CO<sub>2</sub>, and  $H_2S$  in a binary mixture with methane on miscibility using a slim tube apparatus. Sour crude oil from the Comet *T* pool reservoir F Zama field from Alberta, Canada, which contains 3.67%  $CO_2$  and 1.69%  $H_2S$ , was used for these experiments. Slim tube tests were performed at 86.7 °C (188 F) and 17.2 MPa (2500Psig), reflecting the conditions of the reservoir. The results showed that the oil recovery increased with decreasing methane content in the binary mixture. On a comparative basis, the efficiency of  $H_2S$  in reaching miscibility was higher than an equal amount of ethane in the solvent, and the efficiency of  $CO_2$  was lower, while butane and propane have better miscibility than  $H_2S$ .

AlFalahy et al. (1998) summarized the first core flood experiment with H<sub>2</sub>S, which was carried out by Harvey and Henry (1977). The aim of the experiment was to compare the recovery performance of pure  $CO_2$ ,  $H_2S$  and a mixture of them utilizing light and heavy oil samples. Pure H<sub>2</sub>S showed the highest oil recovery in both miscible (light oil) and immiscible conditions (heavy oil). In turn, injection of pure H<sub>2</sub>S resulted in more than 15% more incremental oil recovery, compared to the mixture of H<sub>2</sub>S and CO<sub>2</sub> for both oil samples. CO<sub>2</sub> performed poorly compared to pure H<sub>2</sub>S and mixed gas, but had an advantage over waterflooding in oil recovery. What is more, this work revealed the dependence of the miscibility conditions on the oil composition, as neither H<sub>2</sub>S nor CO<sub>2</sub> injection reached miscibility for heavy oil. Nevertheless, Huerta et al. (2012) reported that the injection of a mixture of 90% CO<sub>2</sub> and 10% H<sub>2</sub>S into heavy oil reservoirs showed a better oil recovery performance than pure CO<sub>2</sub> injection for the cyclic solvent injection process. It was observed that the reduction of oil recovery with subsequent cycles was less by injection CO<sub>2</sub>/H<sub>2</sub>S mixture that demonstrates the benefits of H<sub>2</sub>S content in improving oil recovery over using pure  $CO_2$ .

In Lv et al. (2016) carried out slim tube experiments under reservoir conditions (97 °C) using an oil sample from the K oil reservoir to investigate the displacement efficiency at various displacement pressures and acid gas content. Acid gas was mixed at different concentrations with the hydrocarbon gas. H<sub>2</sub>S and CO<sub>2</sub> constituted 78.11% and 21.89% of the acid gas, respectively. At constant displacement pressure, increasing the acid gas content resulted in a higher displacement efficiency. Under MMP conditions, the displacement efficiency increased linearly with the acid gas composition. Nonetheless, Fig. 11 shows that increasing the acid gas content from 30 to 50% at 25 MPa produced an insignificant improvement in oil recovery, because the estimated MMP values at these acid gas contents were lower than 25 MPa. This observation revealed that, after miscibility is achieved, the effect of increasing the acid gas content on the displacement efficiency is insignificant.

# 3.4. Numerical modeling studies related to sour/acid gas EOR

In addition to experimental studies, efforts have been devoted to numerical modeling of sour gas injection EOR processes compared to other common gases.

AlFalahy et al. (1998) conducted numerical simulations to determine the contribution of  $H_2S$ ,  $CO_2$ , and mixed gas ( $CO_2+H_2S$ ) on oil recovery. Unstable immiscible and stable miscible gas injection schemes were compared utilizing these gases. Pure  $H_2S$  injection resulted in the highest recovery in both cases. In turn, miscible displacement by  $H_2S$  approached 90% oil recovery, followed by its mixture with  $CO_2$ . Compared to that, unstable immiscible conditions resulted in significantly lower recovery, which was around 50% for  $H_2S$  injection. It was concluded that the presence of  $H_2S$  in hydrocarbon gas is beneficial to enhance the oil recovery due to an additional reduction in IFT and oil viscosity. Fig. 12 shows the recovery performance of different gases.

The EOR performance of lean gas, acid gas, sour gas, and  $CO_2$  in low permeable carbonate formation was simulated by Van Vark

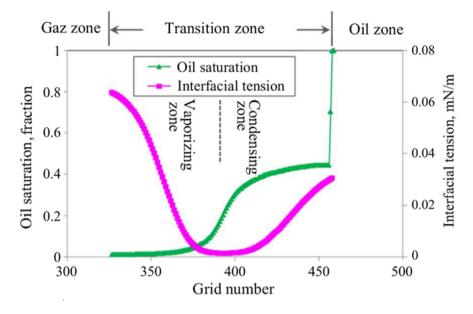


Fig. 9. Interfacial profile within the slim tube (He et al., 2019).

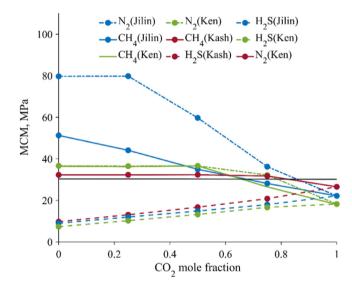


Fig. 10. MCM pressures for various mixtures (Luo et al., 2019b).

et al. (2004). H<sub>2</sub>S, CH<sub>4</sub>, and CO<sub>2</sub> in the sour gas were selected in the 60/30/10 ratio. The acid gas contained  $H_2S$  and  $CO_2$  in the proportion of 3:1. The simulation result showed that the sustainable off-take rates were similar in all gas injection cases, but the plateau rate periods varied due to the different viscosities of the injectants. Sour gas injection resulted in a 25% longer plateau period than lean gas injection, and for more viscous acid gas injection, it was 60%. The same off-take rate indicated that the recovery performance of gas injection depends on the elapsed time and injected gas volume, but not the type of the solvent, although the sweep efficiency becomes stronger with increasing miscibility and seems to be a function of the solvent composition. As acid gas was first contact miscible with oil, while other gases were not, acid gas injection brought about the highest oil recovery. The recovery efficiency of CO<sub>2</sub> lays between acid gas and sour gas as MCM was reached. The gas/oil ratio (GOR) after gas breakthrough increased more slowly during acid gas injection. The authors pointed out that the application of a realistic GOR results in two times more recovery during acid gas injection compared to lean gas injection. Furthermore, the simulation study showed a significant advantage of all gas injection over waterflooding in low permeable (10mD) carbonate reservoirs, despite an unfavorable mobility ratio. Acid gas injection is a more attractive EOR method because of the favorable mobility ratio, better miscibility with oil, and low cost. However, a homogeneous reservoir model was used, so the gas flooding was stable with better sweep efficiency. Therefore, the efficiency of gas injection in heterogeneous reservoirs should be studied further.

Kaita et al. (2020) performed a sensitivity analysis to study the influence of the acid gas composition, gas injection rate, and injection pressure on oil recovery during miscible sour gas injection in Eclipse 300 commercial reservoir simulators. The reservoir model was built based on the one presented by Battistelli et al. (2011). The injectant consisted of 15% acid gas, 80% methane, and 5% hydrocarbon gases. To understand the effect of the acid gas content in the solvent on recovery, various concentrations of acid gas were selected. Moreover, a series of cases were run to compare the recovery performance of pure CO<sub>2</sub>, acid gas (50% H<sub>2</sub>S+50%CO<sub>2</sub>) and sour gas (50% CH<sub>4</sub> and 50% acid gas). The reservoir condition was set slightly higher than MMP for the base case to get the miscible displacement. On a comparative basis, a higher acid gas content in the solvent leads to a higher oil recovery. Increasing the acid gas content from 20% to 40% improves the recovery from 78% to 86%. The higher oil recovery was attributed to a longer plateau period, as the off-take rates for all cases were the same. The increase in acid gas content resulted in greater gas viscosity and better mobility, thus resulting in a longer plateau period and later gas breakthrough. Injection of pure CO<sub>2</sub>, acid gas and sour gas gave similar incremental recovery (around 13%-14%) compared to the base case, and reached 91% after 800 days of production. Similar to the study by Van Vark et al. (2004), the off-take rate decreased with increasing acid gas content, and the plateau rate increased. Despite a similar oil recovery effect, the acid gas is more attractive because of its higher gas viscosity and better mobility, which delay the gas breakthrough. Also, the presence of acid gas decreases the MMP so acid gas injection would be a good method for reviving depleted oil reservoirs.

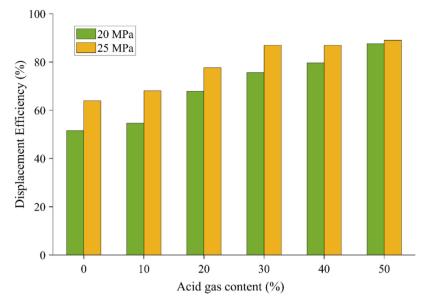


Fig. 11. Displacement efficiency versus acid gas content (Lv et al., 2016).

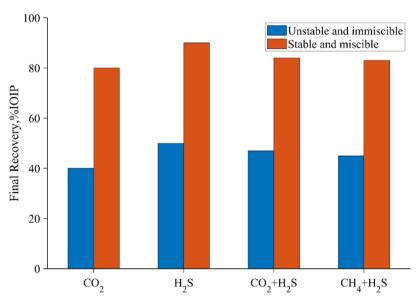


Fig. 12. Oil recovery for injection of various gases (AlFalahy et al., 1998).

# 4. Field experience: sour/acid gas EOR performance

## 4.2. Zama field

# 4.1. Slaughter Estate Unit

One of the first field operations that injected gas containing H<sub>2</sub>S for EOR was performed in the Slaughter Estate Unit (Texas) by Amoco in 1976 (Rowe et al., 1982). A gas mixture with 72% CO<sub>2</sub> and 28% H<sub>2</sub>S was chosen for alternate water and gas injection operation. The acid gas was assumed to have the same displacement efficiency as CO<sub>2</sub> based on laboratory studies. Water alternating gas injection was performed after waterflooding by the simultaneous injection of gas and water at the ratio 1:1. During this project, 26 PV of hydrocarbon gas slugs were injected and then nitrogen was used as the chase gas. The author concluded that the expected incremental tertiary oil recovery resulting from acid gas injection should exceed 20%-25% of the Original Oil in Place, but the final recovery was not discussed. This high incremental oil recovery was used as a key judgment to conclude that MCM occurred in situ. In addition, this operation proved that acid gas with 28%  $H_2S$  was as effective as pure  $CO_2$  injection.

Acid gas (60% CO<sub>2</sub> and 40% H<sub>2</sub>S) injection in the Zama Keg River X2X pool in Alberta, Canada, began in 1995. Davison et al. (1999) reviewed the candidate selection, corrosion monitoring, and key results of this project. They also summarized laboratory studies that were conducted to understand the acid gas miscible displacement, interaction of the acid gas components with brine, the determination of the threshold capillary pressure, and the phase behavior of the injected gas. The results of these studies showed beneficial effects of H<sub>2</sub>S in CO<sub>2</sub>, such as an additional reduction of the MMP and a swelling effect. In 2006, another acid gas injection operation started in Zama Keg River F pool (Smith et al., 2010). The injected acid gas contained 60% CO<sub>2</sub> and 40% H<sub>2</sub>S. By 2011, more than 36,000 metric tons of acid gas had been injected, contributing to 25,000 barrels of incremental oil production. These operations proved the efficiency of acid gas injection as an inexpensive and environmentally friendly means of acid gas disposal and enhancing oil recovery.

#### 4.3. ARF and GAF fields

Masahiro et al. (2002) reviewed the first sour gas injection project in the Gulf Arab region, which was implemented in the oil fields, ARF and GAF, by Abu Dhabi National Oil Company (ADNOC). The project commenced in 2001 and the aim of this project was to reduce the amount of flared acid and sour gas. This was because, before the project, around 7.5 MMSCF/D of acid gas (80% H<sub>2</sub>S and 16% CO<sub>2</sub>) and 2.5 MMSCF/D of sour gas  $(35\% H_2S \text{ and } 2\% CO_2)$  were flared, which had a severe impact on the environment. It was stated that implementation of the sour gas injection project substantially reduced the flared gas volume. The authors explained in detail all the engineering, operational, and economic aspects of this project. However, the review uncovered EOR aspects of this project, which was later discussed by Uchiyama et al. (2008). They stated that high-pressure miscible displacement was achieved by sour gas injection because the reservoir pressure was higher than the dynamic MMP. Monitoring of the produced fluid revealed that the acid gas content increased from 5% up to 15% in the reservoir region where sour gas had been injected, while the injection of sweet gas or sour gas with a lower acid gas concentration reduced the acid content in the produced fluid. However, the composition of the injected sour gas was not included.

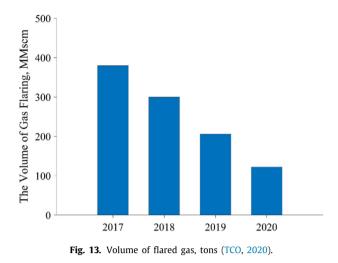
# 4.4. Tengiz oilfield

Tengiz is a huge carbonate sour oil reservoir in the Pre-Caspian basin. The original reservoir oil contains around 15.5% of H<sub>2</sub>S (Bedrikovetsky and Basniev, 1994). The remoteness of the field from the sulfur market and strict environmental regulations in Kazakhstan make sour gas injection into the reservoir attractive. Therefore, the Tengizchevroil (TCO) company began sour gas injection into the Tengiz oil field in October 2007. The central platform was selected for the injection of sour gas due to higher porosity (8%) and fewer fractures compared to the flank side. Sour gas injection in Tengiz is the first contact miscible process and the current reservoir pressure is above the bubble point. As of July 2010, more than 4.3 billion cubic meters of sour gas had been injected into the reservoir, which was approximately 17% of the produced gas. In 2014, the injected sour gas volume reached 14.4 billion cubic meters because of the increasing number of injection wells and injection rate. Sour gas injection in the Tengiz field was considered successful with a high voidage replacement of 110% in 2010 and 250% in 2014, as well as the low ratio between the produced and injected sour gas of less than 1.5% in 2010 and around 3.5% in 2014 (Bealessio et al., 2021). Furthermore, the injection of sour gas resulted in an increase in the reservoir pressure within injection patterns from 47 MPa in 2010 to 54 MPa in 2014. Excellent sweep efficiency - more than 70% for the reservoir part of the injection well - is another measure of success (Darmentaev et al., 2010; Urazgaliyeva et al., 2014).

Sour gas injection significantly reduced the dry gas volume in flaring, which resulted in reduction of the pollutants emission into the air (Byers et al., 2014). Fig. 13 depicts the volume of flared gas by year and shows the gas flaring reduction by 68% in 2020 compared to 2017.

#### 4.5. Kashagan oilfield

Kashagan is a super-giant carbonate oil field in the North Caspian Sea region. The initial reservoir pressure was 78.3 MPa and the reservoir oil contains 18% H<sub>2</sub>S and 5% CO<sub>2</sub> (Akhmedzhanov et al., 2012). Handling the huge volume of sour gas is a big challenge for the operating company. Therefore, as an alternative method of transporting sour gas offshore for treatment, injection



of sour gas into the oil reservoir was launched in December 2012 as part of a pilot program (Galiyeva et al., 2011). Laboratory experiments revealed that oil displacement by the gas is first contact miscible and an insignificant reduction in permeability was observed during the experiment (Ybray et al., 2011). Reservoir simulation till gas breakthrough showed that asphaltene deposits would occupy 5% of the pore space (Malik et al., 2005), which, as the authors mentioned, would not trigger a problem since most deposition occurs near the production well because of a pressure drop. Further monitoring is required to evaluate the feasibility of the sour gas injection project. Table 2 illustrates that the volume of produced gas for reinjection back into the reservoir increased from 28% in 2017 to 41% in 2020. The rest of the gas produced is transported to offshore treatment facilities for export and storage. The increased trend of gas reinjection can be assumed to be a measure of the success of this project.

#### 4.6. Karachaganak gas condensate field

Karachaganak is a giant carboniferous gas condensate reservoir with an initial reservoir pressure of 83-91 MPa. The reservoir mainly contains gas condensate with an underlying oil rim (Ibragimov et al., 2019). The H<sub>2</sub>S and CO<sub>2</sub> composition in the produced fluid is around 3.7% and 5.1%, respectively (Bedrikovetsky and Basniev, 1994). Reinjection of associated gas into Karachaganak gas condensate field was commenced in 2003 and around 40% of the produced gas had been injected into reservoirs to maintain pressure (Kassenov and Kaliyev, 2016).

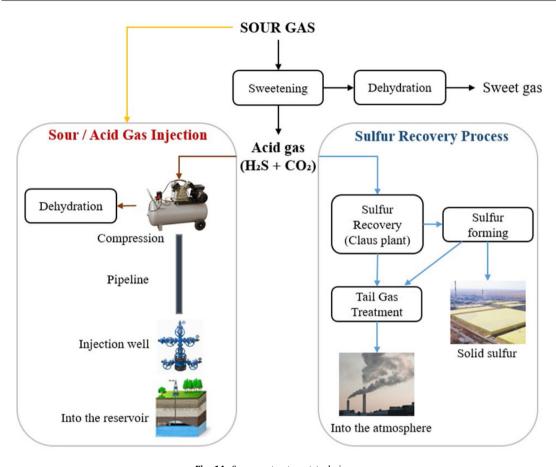
#### 5. Economic analysis

Various methods of handling and utilizing H<sub>2</sub>S from sour gas have been practiced around the world. The technology train of sour gas treatment methods is shown in Fig. 14. The traditional method for managing H<sub>2</sub>S is to separate it from the gas stream and convert it to the elemental sulfur through a number of gas processing and treatment units (Longworth et al., 1996). The main disadvantages of sulfur recovery methods are high investment costs, environmental issues caused by SO<sub>2</sub> and CO<sub>2</sub> emission into atmosphere, and a reduction in sulfur demand in the world (Jamaluddin et al., 1998). The cost-effective alternative for recovering sulfur is to compress the sour/acid gas stream and inject it into the suitable underground formations, which has been successfully implemented in several fields in Canada (Bachu and Gunter, 2005). On the whole, selection and deployment of these methods depend on various economical and technical aspects

#### Table 2

Oil, gas and sulfur production in Kashagan field (NCOC, 2020).

Hydrocarbon and sulfur production	2017	2018	2019	2020
Oil production (wellhead, million tons)	8.29	13.22	14.13	15.14
Gas production (wellhead, billion standard cubic meters)	4.8	7.7	8.45	9.15
Gas reinjection volume (billion standard cubic meters)	0.32	2.24	3.15	3.81
Sulfur production exported (thousand tons)	109	1	1.207	1.228
Sulfur in block storage, year-end (thousand tons)	1.121	1.409	1.527	1.594



**Fig. 14.** Sour gas treatment techniques. *Source:* Adapted from Longworth et al. (1996), Bachu and Gunter (2005).

such as investment and operational cost, sulfur content, sulfur price, and transportation cost. Therefore, managing and handling  $H_2S$  in sour gas requires detailed economic analysis and studies in order to reduce the treatment costs and minimize environmental issues.

Abou-Sayed et al. (2005) discussed the results of the economic analysis of several acid gas management methods on the example of the Tengiz Oilfield. The following acid gas handling methods were considered for the study:

- 1. Solid sulfur storage and disposal at the end of the operation.
- 2. Solid sulfur disposal by injection.
- 3. Acid gas sequestration and disposal.
- 4. Acid gas injection (AGI).

The current practice of sour gas management in the Tengiz Oilfield was selected as a base case for comparison, which involves recovering elemental sulfur from the gas stream and storing it on the ground, while venting the  $CO_2$  into the atmosphere. Economic analysis was carried out to evaluate the impact of recovery, initial investment, and potential regulatory restrictions including taxes for stockpiling the solid sulfur.

Economic value of the first option was calculated without accessing the commercial sale of sulfur. The calculated net present value (NPV) for the first option was -1.9 Billion USD, which includes the cost of the sulfur recovery unit (SRU), operating expense (OPEX) for storing, taxes for stockpiling the sulfur on the ground, and expenses for additional operations to dispose of the sulfur at the end of the project. The NPV of the second option was negative too because of the drilling cost of disposal wells in addition to the capital investment for SRU. Compared to the first option, the economic value of solid sulfur injection is more viable in the long term due to the avoidance of taxation and abandonment liability. The third option of acid gas sequestration in disposal zones has a better economic value than solid sulfur disposal attributing to the elimination of capital investment for sulfur recovery units, taxation and abandonment costs. Furthermore, fewer wells are required for acid gas injection than solid sulfur injection. In the last AGI option, acid gas produced from the sweetening process can be directly injected back into reservoirs, which eliminates the cost of SRUs and storage as well as brings higher incremental oil recovery. Therefore, the AGI option for EOR requires the minimum capital investment while offers the maximum economic return. Overall, the study shows that AGI for

#### Table 3

Advantages and disadvantages of sour gas EOR.

Advantages	Disadvantages
Low MMP; H <sub>2</sub> S storage and utilization;	Potential of $H_2S$ leakage;
low-cost; nearly zero environmental	corrosion of pipes and well
impact; mitigate the risk to people's	abandonment; great caution
health; better sweep efficiency;	needed in terms of safety,
incremental oil production	training, and evacuation

EOR is a superior strategy to others in terms of both economic and environmental concerns (Abou-Sayed et al., 2005).

Several more economic comparisons arrived at similar conclusions. Kokal and Al-Utaibi (2005) evaluated the feasibility of AGI and sulfur recovery for a Saudi Arabia field by discussing the technical and economic factors involved in these processes. Two AGI options were considered, namely pure AGI and slurrified AGI. Results showed that the slurrified AGI is economically unattractive as more wells are required for disposal, while the pure AGI leads to lower capital expenditures (CAPEX) than SRU. However, because of the lower operating costs and the revenues from selling sulfur, it was concluded that SRU is the most favorable option to manage acid gas. But the difference between the NPV values of pure AGI and SRU was small, meanwhile the revenues from incremental oil were not considered. Therefore, pure AGI can be more favorable than SRU in case of additional oil recovery.

Qassim and Mathur (2012) evaluated four options for dealing with acid gas streams after sweetening the produced gas from several sour gas reservoirs in Qatar. The first option was AGI, and the other three considered the construction of a sulfur recovery unit with and without upgrading the existing SRU. All options were evaluated based on the capital expenditures, operating expense, and environmental and safety factors. It was concluded that AGI is a better option than the other three sulfur recovery alternatives because of lower CAPEX and OPEX as well as the minimum impact on the environment. Li et al. (2013) claimed that the reinjection of acid gas requires 40% less investment than the Claus Sulfur recovery option.

Overall, the above studies demonstrated that sour/acid gas injection for EOR is superior to other conventional sour gas treatment techniques in terms of both economic and environmental benefits. The strengths and weaknesses of this method are summarized in Table 3, which are based on the assumption that sour gas is reinjected back into reservoirs, and specialty-designed pipes and equipment for handling the corrosive effects of H<sub>2</sub>S are already installed. In spite of the advantages of sour gas EOR, a detailed economic assessment must be made by taking into account CAPEX, OPEX, possible health, safety, and environmental issues, and technical feasibility before selecting a suitable sour treatment method.

#### 6. Concluding remarks and perspectives

We reviewed the research and application progress of sour gas injection for EOR, which is an economically and environmentally beneficial approach for  $H_2S$  disposal, as well as an effective technique for improving oil recovery. Previous laboratory experiments and simulation studies demonstrated the superiority of sour gas injection to  $CO_2$  or light hydrocarbon gas injection from the viewpoint of oil recovery.

- H<sub>2</sub>S has a stronger miscible ability than methane and CO<sub>2</sub> with crude oil. Hence, sour gas injection is more apt to achieve miscibility under similar operating conditions, which is a function of H<sub>2</sub>S concentration.
- The contribution of H<sub>2</sub>S to the reduction of MMP is greater than that of CO<sub>2</sub> in hydrocarbon gas. Thus, the presence of H<sub>2</sub>S would be beneficial for CO<sub>2</sub> injection because of additional MMP reduction.

- The phase envelope of crude oil becomes narrower after the injection of sour gas. The bubble point pressure decreases due to H<sub>2</sub>S, which suppresses the liberation of the solution gas and maintains a single-phase flow in the reservoirs.
- Addition of H<sub>2</sub>S to hydrocarbon gas raises the density and viscosity of the gas mixture, which improves mobility control during immiscible gas flooding and prevents early gas breakthrough in a multiphase flow scenario.
- Sour gas injection is an economically favorable technique for handling and utilizing H<sub>2</sub>S because of lower capital investment and operational expenses as well as incremental oil recovery compared to other sour gas management methods.

Although multiple studies have been conducted to investigate the effectiveness of sour gas injection EOR, systematic knowledge and guidelines on the design and optimization of the processes are still limited, due to the reality that sour gas injection in the field is implemented primarily for H<sub>2</sub>S disposal. A few directions that deserve better understanding are listed below:

- The phase behavior alteration of crude oil caused by sour gas injection needs more experimental and numerical efforts for complete and in-depth illustration. The toxic nature of H<sub>2</sub>S restrains experimental studies due to the serious precautions and safety measurements. In recent years, the increasingly mature microfluidics are opening up new opportunities for sour gas injection experiments, in which a small dosage of H<sub>2</sub>S is required, significantly reducing the safety concerns associated with H<sub>2</sub>S.
- In carbonate reservoirs, the interactions of the injected sour gas with brine and rock require extensive experimental and modeling studies, which have so far been rarely touched on.
- Sector and reservoir scale modeling of sour gas injection for EOR has not been fully developed or at least reported. To explore the features of sour gas injection, for example better mobility control; it is necessary to conduct simulations using realistic geological models with heterogeneity.
- Currently, machine learning (ML) methods are believed to be an effective alternative to conventional field-scale reservoir simulations and modeling techniques for reducing the computational cost and improving the integrity of predicting the efficiency of various EOR processes (You et al., 2020); (Vo Thanh et al., 2020). Nonetheless, ML techniques have not been used for developing robust and fast predictive models to evaluate the sour gas injection for EOR. Therefore, construction of ML proxy models for reducing uncertainties associated with insufficient laboratory data is one of the future research directions.
- As can be seen from the literature review above, there are still some ambiguities with sour gas flow in porous media and the geochemical interaction of H<sub>2</sub>S with the formation brine and rocks during sour gas injection. Hence, more studies should be conducted to investigate the sour gas flow in porous media, including fractured carbonate reservoirs, and H<sub>2</sub>S-induced mineral dissolution, which may alter reservoir porosity and permeability, using reactive transport modeling, such as the lattice Boltzmann method (Kang et al., 2010; Tian et al., 2016).

# Nomenclature

AGI Acid gas injection CAPEX Capital expenditure EOR Enhanced oil recovery EoS Equation of state FCM First-contact miscibility GOR Gas/oil ratio IFT Interfacial tension MCM Multi-contact miscibility MMP Minimum miscibility pressure MW Molecular weight NPV Net present value OPEX Operating expense PPM Parts per million PV Pore volume PVT Pressure-volume-temperature RBA Rising bubble apparatus SRU Sulfur recovery unit

# CRediT authorship contribution statement

**Madiyar Koyanbayev:** Conceptualization, Investigation, Compilation, Writing – original draft. **Lei Wang:** Conceptualization, Supervision, Writing – review and editing. **Yanwei Wang:** Conceptualization, Supervision, Writing – review and editing. **Muhammad Rehan Hashmet:** Conceptualization, Supervision, Writing – review and editing.

## **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. **Data availability** 

Data will be made available on request.

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