Improvement of Hydrogen Sulfide Scavenging via the Addition of Monoethanolamine to Water-Based Drilling Fluids

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ABSTRACT: The release of the lethal hydrogen sulfide (H₂S) gas during the drilling of sour subterranean formations is of huge health and safety concern. Additionally, the contact of this corrosive gas with handling equipment might result in severe damages and significant economic losses. Accordingly, effective in situ scavenging of H₂S while drilling is very crucial. Thus, we report herein the addition of monoethanolamine (MEA) to water-based mud with the objective of improving the H₂S-scavenging efficacy of the mud. The H₂S-scavenging capacity was evaluated for the MEA-containing mud and compared with the base mud and fluids containing the commercial scavengers, SourScav and triazine. Also, the key mud characteristics including rheology, filtration properties, alkalinity, and corrosion rate were investigated in the presence and the absence of MEA, and the obtained results were compared to those of SourScav and triazine. The obtained experimental results revealed that the addition of MEA to the base mud significantly improved the H₂S adsorption capacity of the base mud by 117%, compared to 50 and 74% with the SourScav and triazine. Additionally, the pH value of the MEA-containing mud complied with the practical recommendations for drilling in a sour environment. Moreover, the plastic viscosity was increased by 13% to 37 cP with the MEA-containing mud with an insignificant impact on the yield point. Furthermore, the MEA-containing mud showed a favorable zero corrosion rate, as was the case for SourScav and triazine muds. However, unlike SourScav and triazine, MEA did not substantially enhance the filtration performance. Nonetheless, the filtration characteristics of the MEA-containing mud were still better than the base mud and within the recommended practical range for water-based muds. Overall, the results presented in this study reveal that the addition of monoethanolamine to water-based muds improves the mud characteristics and, more importantly, provides a superior H₂S-scavenging performance relative to SourScav and triazine scavengers, suggesting the commercial relevance of MEA for drilling applications.

1. INTRODUCTION

In oil and gas drilling, each additive has a certain role to play. Thus, the appropriate selection of additives and the proper design of drilling muds are critical factors for successful oil and gas drilling operations.1−4 The drilling mud can be categorized as aqueous, nonaqueous, and gaseous-based mud. The aqueous mud (i.e., water-based mud) is the most commonly used in drilling operations with preferable technical, commercial, and environmental attributes.5−7

The release of hydrogen sulfide gas (H₂S) during the drilling of sour subterranean formations is a commonly encountered problem. H₂S, also known as hydrosulfuric acid or sewer gas, is the most encountered sulfhydryl compound, in addition to mercaptans, thiol carboxylic acids, and dithio acids. It is a highly corrosive, poisonous, colorless, flammable, reactive, and heavy (with a molecular weight of 34.08 and specific gravity of 1.18 relative to air) gas.8 H₂S is the most reduced form of sulfur, and it produces the distinctive odor of rotten eggs, which can be detected by smell at concentrations as low as 0.5 ppm (part per million). H₂S naturally exists in oil and gas reservoirs, originating from geological sources or microbiological processes.9−12 H₂S can invade the drilling fluid system during the drilling operations from either the metabolism of sulfate-reducing bacteria (SRB) that grow in anaerobic oilfield conditions, the thermal decomposition of sulfur-containing drilling mud additives, or by invading H₂S pockets or formations containing underground water and oil that are contaminated with this gas.13−16

1.1. Impacts of H₂S on Drilling Operations. H₂S is encountered during drilling operations in various regions of the world, including the United States, Canada, Venezuela, Russia, China, and Middle Eastern countries such as Saudi Arabia, Iraq, Oman, Syria, Egypt, and Iran.17−19 Significant amounts of H₂S
Table 1. Types of \( \text{H}_2\text{S} \) Scavengers Used in Drilling Operations

| Type                        | Chemical Reaction                                                                 | Remarks                                                                 |
|-----------------------------|-----------------------------------------------------------------------------------|------------------------------------------------------------------------|
| oxidizers (e.g., hydrogen peroxide, \( \text{H}_2\text{O}_2 \), and potassium permanganate, \( \text{KMnO}_4 \)) | \( \text{H}_2\text{O}_2 + \text{H}_2\text{S} \rightarrow \text{S}^{\text{2-}} + 2\text{H}_2\text{O} \) | \( \text{H}_2\text{O}_2 \) is a nonselective reactant                   |
| copper compounds (e.g., copper carbonate, \( \text{CuCO}_3 \), and copper nitrate, \( \text{Cu(NO}_3)_2 \)) | \( 8\text{KMnO}_4 + 3\text{H}_2\text{S} \rightarrow 3\text{K}_2\text{SO}_4 + 8\text{MnO}_2 + 2\text{KOH} + 2\text{H}_2\text{O} \) | uncontrollable scavenging process                                   |
|                         | \( \text{CaCO}_3 + \text{H}_2\text{O} \rightarrow \text{CaS} + 2\text{H}_2\text{O} \) | not effective with heavy mud weight                                   |
|                           | \( \text{Cu(NO}_3)_2 + \text{H}_2\text{S} \rightarrow \text{CuS} + 2\text{HNO}_3 \) | metal-based scavenger                                                 |
|                           | \( \text{ZnO} + \text{H}_2\text{S} \rightarrow \text{ZnS} + 2\text{H}_2\text{O} \) | efficient and fast reaction                                           |
|                           | \( \text{Fe(C}_6\text{H}_4\text{O}_7)_3 + \text{H}_2\text{S} \rightarrow \text{FeS} + 2\text{C}_6\text{H}_4\text{O}_7 + \text{H}_2\text{O} \) | corrosion from copper electrodeposition                                |
|                           | \( \text{H}_2\text{O}_2 + \text{H}_2\text{S} \rightarrow \text{S}^{\text{2-}} + 2\text{H}_2\text{O} \) | enhanced fluid rheology and filtration properties                      |
| zinc compounds (e.g., zinc oxide, \( \text{ZnO} \)) | \( \text{ZnO} + \text{H}_2\text{S} \rightarrow \text{ZnS} + 2\text{H}_2\text{O} \) | they have amphoteric nature with predictable reaction and thermal stability |
| iron compounds (e.g., iron oxide, \( \text{Fe}_2\text{O}_3 \), and iron gluconate, \( \text{Fe(C}_6\text{H}_4\text{O}_7)_3 \)) | \( \text{Fe}_2\text{O}_3 + 3\text{H}_2\text{S} \rightarrow 3\text{FeS} + 2\text{H}_2\text{O} \) | higher amounts deteriorate the mud rheology and cause flocculation and fluid losses at high pH |
|                            | \( \text{Fe(C}_6\text{H}_4\text{O}_7)_3 + \text{H}_2\text{S} \rightarrow \text{FeS} + 2\text{C}_6\text{H}_4\text{O}_7 + \text{H}_2\text{O} \) | active and magnetic iron oxides are in use                            |
|                            | \( \text{Fe}_2\text{O}_3 + 6\text{H}_2\text{O} \rightarrow 2\text{FeS} + 4\text{H}_2\text{O} \) | they increase the mud density                                         |
|                            | \( \text{Fe}_2\text{O}_3 + 6\text{H}_2\text{O} \rightarrow 2\text{FeS} + 4\text{H}_2\text{O} \) | more effective at low pH                                              |
|                            | \( \text{Fe}_2\text{O}_3 + 6\text{H}_2\text{O} \rightarrow 2\text{FeS} + 4\text{H}_2\text{O} \) | \( \text{Fe}_2(\text{C}_6\text{H}_4\text{O}_7)_3 \) is an eco-friendly scavenger with a fast reaction |

1.2. \( \text{H}_2\text{S} \) Scavengers Used with Drilling Fluids. Due to its associated impacts and damages, \( \text{H}_2\text{S} \) is one of the hazardous gases associated with oil and natural gas. Therefore, the safe and immediate tackling of any released \( \text{H}_2\text{S} \) during the drilling operations is a must. A variety of specialized additives or chemicals were applied in drilling operations to selectively react and effectively scavenge \( \text{H}_2\text{S} \) without producing any unwanted byproducts. Since \( \text{H}_2\text{S} \) reacts with strong oxidizers, concentrated nitric acid, and metals, most of the used \( \text{H}_2\text{S} \) removal approaches are based either on ionic precipitation or surface adsorption. Table 1 describes the \( \text{H}_2\text{S} \) scavengers used in drilling operations.

Although several scavengers were presented and applied in the oil and gas industry, each one has its advantages and limitations. Generally, the limitations can be related to reactivity and kinetics, scavenging mechanism, conditions, cost, or/hand HSE (health, safety, and environmental) issues. Accordingly, research is still ongoing to develop an ideal scavenger that possesses a complete and fast \( \text{H}_2\text{S} \) removal without producing undesirable byproducts. The developed scavenger should be feasible for applications in the petroleum industry.

Alkanolamines, like monoethanolamine (MEA), diethanolamine (DEA), and \( N \)-methyl diethanolamine (MDEA), are commonly used to sweeten sour gases in stream facilities. These scavengers are regenerative, enabling their repetitive use. However, these compounds are employed mainly in sour gas sweetening with controlled process parameters such as the amine concentrations, inlet gas temperature, pressure, circulation rate, and the number of contact stages to minimize operational problems.

MEA was also utilized in the literature to increase the thermal stability of starch polymer in water-based mud. The results indicated the successful improvement in starch stability as MEA helped to achieve the functional role of the starch polymer by reducing the filtration characteristics and increasing the viscosity. Some other applications of MEA include cosmetics, cleaning products, pH controllers, corrosion inhibitors, plasticizing agents, agricultural sprays, emulsion paints, and pharmaceuticals.
To the best of our knowledge, no previous studies have been performed to evaluate the usage of MEA as an H2S scavenger with drilling fluids. Therefore, this work aims to assess the effects of adding MEA to water-based drilling muds in terms of H2S-scavenging improvement and the alteration (if any) of other mud properties (i.e., alkalinity, rheology, and filtration performance). Additionally, the corrosion rates of the prepared drilling fluids were evaluated and compared.

2. MATERIALS

A mud recipe from field applications was used to prepare 350 cm³ of water-based drilling fluid samples. The base fluid was fresh water, and some functional additives were added sequentially under ambient conditions using a high-speed Hamilton Beach mixer. The practical additives were mixed to maintain the viscosity, alkalinity, fluid loss, shale swelling, and filtration. The barite quantity of 150 g was used to obtain 1,486 g/cm³ (12.4 ppg) mud weight. Each H2S scavenger was added at the end and mixed for 10 min. Table 2 shows the drilling fluid formulation stating the mixing sequence, quantities, mixing time, and function of each component.

Table 2. Formulation of Base and H2S Scavenger-Containing Muds

| component            | quantity | mixing duration (min) | function            |
|----------------------|----------|-----------------------|---------------------|
| water                | 308 cm³  | base fluid            |                     |
| defoamer             | 0.08 cm³ | 1                     | antifoam agent      |
| xanthan gum          | 1 g      | 20                    | viscosity controller|
| starch               | 6 g      | 15                    | fluid loss controller|
| PAC-R                | 1.5 g    | 15                    | fluid loss controller|
| NaCl                 | 60 g     | 10                    | shale stabilizer    |
| caustic soda         | 0.5 g    | 1                     | alkalinity controller|
| CaCO3                | 15 g     | 10                    | bridging material   |
| barite               | 150 g    | 10                    | weighting agent     |
| H2S scavenger        | 0/1 g    | 10                    | H2S scavenger       |

Two commercial H2S scavengers (i.e., triazine and SourScav), supplied by a drilling fluid services company, were used as references for comparison with the proposed scavenger. The obtained triazine is in the liquid phase with a density of 1.073 g/cm³, pH of 10.5, and average concentration of 60 vol %. While SourScav is an iron gluconate-based powder that is soluble in water and has a density of 0.7 g/cm³, triazine, and MEA. The test setup is described in detail thereafter. Two commercial H2S scavengers (i.e., triazine and SourScav), supplied by a drilling fluid services company, were used as references for comparison with the proposed scavenger. The obtained triazine is in the liquid phase with a density of 1.073 g/cm³, pH of 10.5, and average concentration of 60 vol %. While SourScav is an iron gluconate-based powder that is soluble in water and has a density of 0.7 g/cm³, pH of 4.5, and average concentration of 60 vol %. The used MEA (C2H4NO) is an organic viscous and colorless liquid that is miscible with water and has an ammonia-like odor. It has a density of 1.01 g/cm³ and pH of 12.1 and is usually produced from the reaction of ethylene oxide with ammonia.

3. EXPERIMENTAL WORK

Several laboratory tests were conducted in this study to thoroughly investigate the applicability of MEA with water-based mud. The methodology is summarized in Figure 1 and described in detail thereafter.

After preparing the drilling fluid samples, the H2S-scavenging experiments were conducted at ambient conditions to assess the H2S-scavenging capacity of the base mud and fluids containing SourScav, triazine, and MEA. The test setup is described in Figure 2. In this experiment, 10 cm³ from each mud sample was placed in a burette with an inlet of gas from a cylinder containing 100 ppm H2S, while the burette outlet is connected to a MultiRAE gas detector having a minimum detection limit of 0.1 ppm. A flowmeter was used at the gas inlet to control the gas flow rate at 150 cm³/min. The outlet gas concentration was continuously recorded until it reached the maximum (saturation) concentration of 100 ppm (i.e., equal to the inlet gas concentration).

The H2S-scavenging capacity (in mg H2S/l mud) by each mud formulation when the scavenger was fully consumed (i.e., saturation was attained) was calculated using the following equation:

\[
saturatio\text{ }cain\text{ }ouc\text{ }y (\text{mg/L}) = 150 \times 10^{-7} \times \rho \times \int_0^{t_s} (100 - C_{out})\,dt
\]  

where \(\rho\) is the H2S density (1.391 mg/cm³), \(t_s\) is the saturation time in minutes at the outlet H2S concentration of 100 ppm, and \(C_{out}\) is the H2S concentration at the outlet gas stream.

The pH value of each mud was measured using a pH meter at ambient conditions.

The rheology measurements were conducted according to the American Petroleum Institute (API) standard procedures. The measured rheological properties included plastic viscosity (PV), yield point (YP), and gel strengths at 10 s and 10 min. The OFITE viscometer model 900 was used to perform these measurements at 120 °F. The values of PV (cp) and YP (lb/100 ft²) were calculated from the shear stress–shear rate relationship using the Bingham plastic model. Additionally, the gel strength values were measured by momentarily stirring the fluid sample at a low shear rate (i.e., 3 rpm) after staying static for 10 s and 10 min. These rheological measurements were performed for the base, reference, and MEA-containing muds.

The filtration test was also performed for all muds following the API standards and using the high-pressure high-temperature (HPHT) OFITE filter press to address the filtration conduct. The test was carried out using a 40 μm ceramic disk as the filtration media under 300 psi of differential pressure and a temperature of 250 °F. The volume of the filtrated fluid was recorded for 30 min and collected in a graduated cylinder. Then, the thickness and weight of the formulated filter cake were measured.

The corrosion rate for the MEA-containing fluid was evaluated and compared to those of the base and reference fluids. The HPHT corrosion test was conducted using metal coupons, from casing steel of N80 grade, soaked in the prepared muds for 6 h at 250 °F and 300 psi inside anticrosive autoclave cells.

4. RESULTS AND DISCUSSION

4.1. H2S-Scavenging Test. The results of the scavenging test showed (see Figure 3) that H2S started to breakthrough after 8 min and reached the saturation concentration after 85 min of contact with the base mud. Using SourScav helped to postpone the H2S breakthrough and saturation times till after 13 and 120 min, respectively. Triazine, on the other hand, postponed the H2S breakthrough and saturation times to 51 and 133 min, respectively. Adding MEA to the base drilling mud improved the fluid-scavenging capacity by delaying the breakthrough and saturation times up to 35.5 and 176 min, respectively.

The H2S-scavenging capacities at the saturation condition were calculated using eq 1. The obtained results indicated that the base mud had a capacity of 125 mg of scavenged H2S/L of mud, whereas the commercial SourScav enhanced the saturation.
capacity by 50% to 187 mg/L and the triazine improved the saturation capacity of the drilling fluid by 74% to reach 217 mg/L. On the other hand, the addition of MEA to the base mud resulted in a saturation capacity of 270 mg/L, which is equivalent to a 117% improvement over the base mud, revealing the outstanding performance of MEA (Figure 4). The results also demonstrate that the MEA-scavenging capacity at the saturation condition outperforms triazine and SourScav.

The applied commercial scavengers are iron gluconate- and triazine-based products; therefore, the presence of these compounds (i.e., iron gluconate and triazine) contributed to the above-described scavenging performance. The reaction chemistry for MEA is designed to reduce the H₂S concentrations to minimum levels through the neutralization reaction and producing a sulfide derivative. It is worth mentioning that MEA is characterized as a regenerative scavenger and stable...
compound with no thermal decomposition or degradation up to its normal boiling point (338 °F). It has a low molecular weight, which results in high solution capacity at low to moderate concentrations. Also, its high pH value and its relatively simple recovery process from contaminated solutions are advantageous. However, the most serious disadvantage is the irreversible reaction when the system temperature is heated to ∼245 °F at 10 psig, as per the below reaction

\[
\text{C}_2\text{H}_5\text{NO} + \text{H}_2\text{S} \rightarrow \text{C}_2\text{H}_5\text{NOH}^+ + \text{HS}^-
\]

The H₂S-scavenging performance of MEA is attractive. However, to assess its suitability for drilling applications, its impact on mud properties (i.e., rheology, alkalinity, corrosion rate, and filtration) must be also evaluated. Accordingly, these properties were investigated, and the obtained results are presented and discussed below.

### 4.2. Rheology Test

The rheology measurements indicated that the base mud had a PV value of 32.5 cP and YP of 95.9 lb/100 ft² with 10 s/10 min gel strengths of 6/7 lb/100 ft². Adding the SourScav decreased both the plastic viscosity and the yield point to 25.8 cP and 72.8 lb/100 ft², respectively. While triazine reduced the PV to 29.9 cP but preferably increased the YP to 108.8 lb/100 ft². The proposed material, MEA, increased the PV to 37 cP with only a 7% decrease in the YP of the fluid (Figure 5).

The increment in PV accompanied by an insignificant decrease in the YP is in accordance with the previous study on the MEA effect on mud rheology. On the other hand, the 10 s/10 min gel strengths were dropped to 5/6 and 4/5 lb/100 ft² with MEA and SourScav, respectively, which would help in lowering the equivalent circulating density, while the gel strengths were increased to 7/8 lb/100 ft² with triazine (Figure 6a). The performance of shear stresses at low shear rates for the examined fluid samples, which are depicted in Figure 6b, can justify the obtained strength values.

### 4.3. pH Measurement

The pH measurements provided pH values of 11.4, 9.1, 11.5, and 11.6 for the base, SourScav, triazine, and MEA-containing muds, respectively (Figure 7).

The lower pH of SourScav compared to the higher pH of both triazine and MEA is attributed to the significant pH change induced by the scavengers. Generally, the practical pH range of water-based drilling muds is in the range of 9.0–11.0; however, increasing the mud pH value above this range is a recommended practice to limit the H₂S effects, and the regulations set pH level of 10 to be always maintained as a minimum in a sour environment. Accordingly, the pH resulting from the use of the MEA complies with the regulations and recommendations.

### 4.4. Corrosion Test

The results from corrosion tests showed that the base mud had a corrosion rate of 0.9 × 10⁻⁵ lb/ft², whereas using the reference muds (SourScav and triazine) gave almost zero corrosion rate. Also, a zero corrosion rate was observed for the MEA-containing mud after 6 h of contact between the mud formulations and the coupon, revealing the noncorrosive nature of MEA. The limited MEA solution...
concentration helps to avoid the corrosion problem that arises at concentrations exceeding 20%. Moreover, MEA is used to prevent corrosion in oil well chemicals and metalworking, which confirms the obtained results.\textsuperscript{72,75}

4.5. Filtration Test. The filtration test for the base mud revealed that the filtrated volume was 11.8 cm\textsuperscript{3} and the formulated filter cake had thickness and weight of 3 mm and 22 g, respectively. The addition of SourS\textsuperscript{2}c\textsubscript{2} and triazine significantly enhanced the filtration performance with less filtrated volume and better filter cake features, as shown in Figure 8. The MEA addition to the base mud resulted in a slight decrease in the filtrated volume to 11.6 cm\textsuperscript{3}. Additionally, the thickness and weight of the composed filter cake were also decreased to 2.9 mm and 21 g, respectively. Although the MEA did not substantially enhance the filtration behavior, the resulted characteristics are still better than the base mud and within the practical range for water-based muds. These results indicated a better plugging mechanism that could lead to reduced formation damage.

This study shows that MEA can be successfully added to water-based muds to effectively scavenge hydrogen sulfide. Additionally, the MEA-containing mud possesses competent mud properties. However, further research and optimization of the mud formulation are still required before field application trials.

5. CONCLUSIONS

In this work, MEA was introduced as an H\textsubscript{2}S scavenger to enhance the scavenging capacity of water-based muds. The obtained scavenging capacity and effects on mud properties were compared to those of the base, SourS\textsuperscript{2}c\textsubscript{2}, and triazine muds with the following conclusions:

- Using MEA considerably improved the H\textsubscript{2}S-scavenging capacity by 117% relative to the base mud, while the commercial SourS\textsuperscript{2}c\textsubscript{2} and triazine increased the capacity by 50 and 74%, respectively. Additionally, the MEA-containing mud had a pH value that complies with the practical recommendations for drilling in a sour environment.
- MEA increased the plastic viscosity of the mud from 32.5 to 37 cP with an insignificant impact on the yield point.
- The resulted zero corrosion rate after 6 h of contact between the MEA-containing mud and the metal coupon indicated the noncorrosive nature of the MEA; SourS\textsuperscript{2}c\textsubscript{2} and triazine also displayed noncorrosive behavior.
- Adding MEA to the base drilling mud slightly enhanced the filtration performance; however, SourS\textsuperscript{2}c\textsubscript{2} and triazine provided preferable filtration characteristics.
- Further research and optimization of the mud formulation are required before field application trials.

8. Results of the filtration test.

Figure 8. Results of the filtration test.

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