

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

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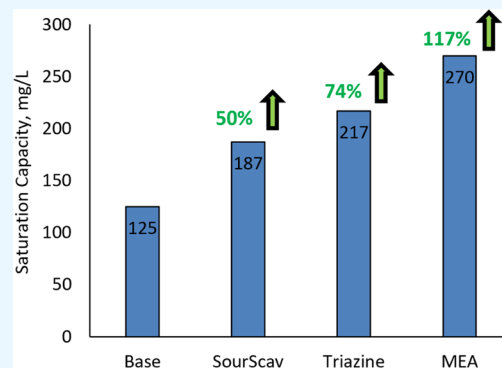
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ABSTRACT: The release of the lethal hydrogen sulfide (H_2S) gas during the drilling of sour subterranean formations is of huge health and safety concern. Additionally, the contact of this corrosive gas with handling equipment might result in severe damages and significant economic losses. Accordingly, effective in situ scavenging of H_2S while drilling is very crucial. Thus, we report herein the addition of monoethanolamine (MEA) to water-based mud with the objective of improving the H_2S -scavenging efficacy of the mud. The H_2S -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 H_2S 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 H_2S -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 (H_2S) during the drilling of sour subterranean formations is a commonly encountered problem. H_2S , also known as hydrosulfuric acid or sewer gas, is the most encountered sulfhydryl compound, in addition to mercaptans, thiol carboxylic acids, and dithio acids.⁸ 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.⁹ H_2S 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). H_2S naturally exists in oil and gas reservoirs, originating

from geological sources or microbiological processes.^{10–12} H_2S 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 H_2S pockets or formations containing underground water and oil that are contaminated with this gas.^{13–16}

1.1. Impacts of H_2S on Drilling Operations. H_2S 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 H_2S

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Table 1. Types of H₂S Scavengers Used in Drilling Operations

type	chemical reaction	remarks
oxidizers (e.g., hydrogen peroxide, H ₂ O ₂ , and potassium permanganate, KMnO ₄) ^{11,12,23,38–54}	$\text{H}_2\text{O}_2 + \text{H}_2\text{S} \rightarrow \text{S}^{2-} \downarrow + 2\text{H}_2\text{O}^+$	–H ₂ O ₂ is a nonselective reactant –uncontrollable scavenging process
	$8\text{KMnO}_4 + 3\text{H}_2\text{S} \rightarrow 3\text{K}_2\text{SO}_4 + 8\text{MnO}_2 + 2\text{KOH} + 2\text{H}_2\text{O}$	–KMnO ₄ showed preferred mud rheology –not effective with heavy mud weight –metal-based scavenger
copper compounds (e.g., copper carbonate, CuCO ₃ , and copper nitrate, Cu(NO ₃) ₂) ^{36,37,41,44,46–49}	$\text{CuCO}_3 + \text{H}_2\text{S} \rightarrow \text{CuS} \downarrow + \text{H}_2\text{O} + \text{CO}_2 \uparrow$	–efficient and fast reaction –corrosion from copper electrodeposition
	$\text{Cu}(\text{NO}_3)_2 + \text{H}_2\text{S} \rightarrow \text{CuS} \downarrow + 2\text{HNO}_3$	–Cu(NO ₃) ₂ showed less affinity to corrosion –enhanced fluid rheology and filtration properties –they have amphoteric nature with predictable reaction and thermal stability
zinc compounds (e.g., zinc oxide, ZnO) ^{14,37,46,50–58}	$\text{ZnO} + \text{H}_2\text{S} \rightarrow \text{ZnS} \downarrow + \text{H}_2\text{O}$	–higher amounts deteriorate the mud rheology and cause flocculation and fluid losses at high pH
	$\text{Fe}_3\text{O}_4 + 6\text{H}_2\text{S} \rightarrow 3\text{FeS}_2 \downarrow + 4\text{H}_2\text{O} + \text{H}_2$	–active and magnetic iron oxides are in use –they increase the mud density
iron compounds (e.g., iron oxide, Fe ₃ O ₄ , and iron gluconate, Fe(C ₆ H ₁₂ O ₇) ₂) ^{30,37,39,48,59–66}		–more effective at low pH
	$\text{Fe}(\text{C}_6\text{H}_{12}\text{O}_7)_2 + \text{H}_2\text{S} \rightarrow \text{FeS} \downarrow + 2\text{C}_6\text{H}_{12}\text{O}_7 + \text{H}_2\text{O}$	–Fe(C ₆ H ₁₂ O ₇) ₂ is an eco-friendly scavenger with a fast reaction –resulted pH drop –not effective with heavy mud weight

might be released from these operations, posing serious health and safety challenges. The release of H₂S also results in sour corrosion to equipment, which occurs because of either increasing the contained-fluid acidity or forming a corrosive iron sulfide.^{8,12,20–25} The formed corrosive iron sulfide strongly adheres to steel surfaces as a scale and may result in pitting corrosion, which shortens the equipment life and can precipitate and result in the formation of plugging.^{26,27} H₂S may also react with steel as a catalyst or accelerator to produce hydrogen ions, which results in metal brittle failure through either hydrogen embrittlement or stress cracking.^{16,24,28,29}

When H₂S is transferred to the drilling fluid, it affects the mud viscosity, fluid loss, and density, causing a problem of well kick, in addition to the reduction in pH, making the mud more corrosive to metallic components.^{12,23,30,31} It is worth mentioning that H₂S liquefies at as low pressures as 350–400 psi. Therefore, during the H₂S-containing kick, H₂S stays in the liquid phase till it reaches close to the surface, where it will be transferred to the gaseous phase with a sudden and huge increase in volume, resulting in a challenging well-control situation.¹⁶

Moreover, the escape of this lethal gas to the surface poses extreme hazards to the health of personnel depending on the duration and level of exposure. Because the human nose is extremely sensitive to H₂S odor, even minute amounts of this gas may be recognized. Most people can perceive the first detectable rotten egg odor of H₂S at concentrations ranging from 0.01 to 1.5 ppm, and the odor gets more offensive at concentrations ranging from 3 to 5 ppm. However, take caution when relying on the sense of smell to identify H₂S since larger amounts of H₂S anesthetize the smell sense.¹⁶ The standard regulations and recommendations for the H₂S exposure limit are developed and enforced by the relevant agencies, such as the Occupational Safety and Health Administration (OSHA) and National Institute for Occupational Safety and Health (NIOSH), with regular updates. NIOSH recommends a 10 min threshold limit of exposure to 10 ppm and identifies a concentration of 100 ppm as an immediate life-threatening level.^{9,16,32–34}

1.2. H₂S Scavengers Used with Drilling Fluids. Due to its associated impacts and damages, H₂S is one of the hazardous gases associated with oil and natural gas. Therefore, the safe and immediate tackling of any released H₂S during the drilling operations is a must.^{35,36} A variety of specialized additives or chemicals were applied in drilling operations to selectively react and effectively scavenge H₂S without producing any unwanted byproducts. Since H₂S reacts with strong oxidizers, concentrated nitric acid, and metals, most of the used H₂S removal approaches are based either on ionic precipitation or surface adsorption.^{12,37} Table 1 describes the H₂S 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/and HSE (health, safety, and environmental) issues. Accordingly, research is still ongoing to develop an ideal scavenger that possesses a complete and fast H₂S removal without producing undesirable byproducts. The developed scavenger should be feasible for applications in the petroleum industry.³⁷

Alkanolamines, like monoethanolamine (MEA), diethanolamine (DEA), and *N*-methyl diethanolamine (MDEA), are commonly used to sweeten sour gases in stream facilities. These scavengers are regenerable, 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.⁶⁷

MEA was also utilized in the literature to increase the thermal stability of starch polymer in water-based mud.⁶⁸ 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.^{69–71}

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 cm ³		base fluid
defoamer	0.08 cm ³	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
CaCO ₃	15 g	10	bridging material
barite	150 g	10	weighting agent
H ₂ S scavenger	0/1 g	10	H ₂ S 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, pH of 4.5, and average concentration of 60 vol %. The used MEA (C₂H₇NO) 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 equation⁴⁵

$$\begin{aligned} \text{saturation capacity (mg/L)} \\ = 150 \times 10^{-7} \times \rho \times \int_0^{t_s} (100 - C_{\text{out}}) dt \end{aligned} \quad (1)$$

where ρ 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 and 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 anticorrosive 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

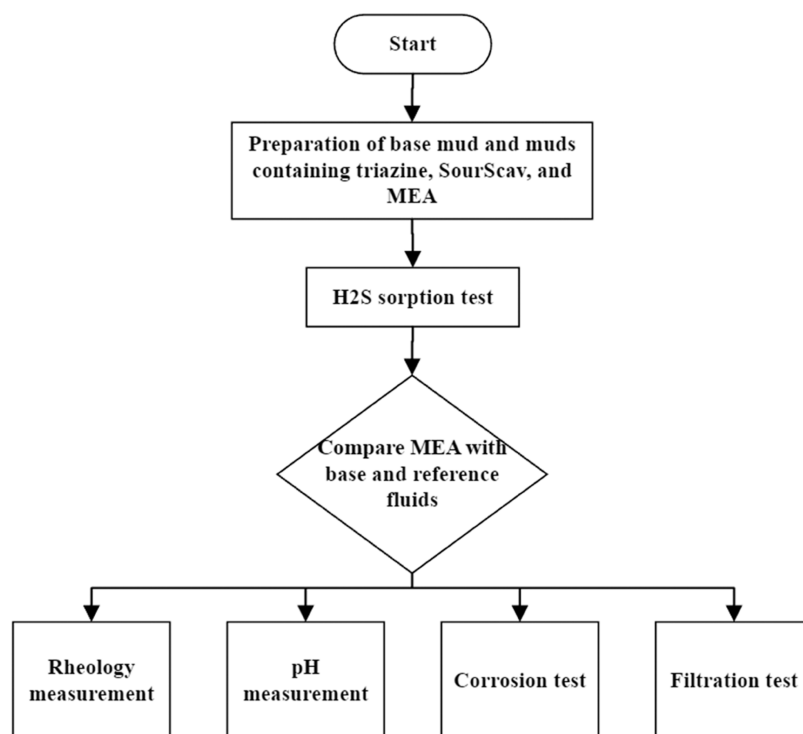


Figure 1. Flowchart of the experimental work.

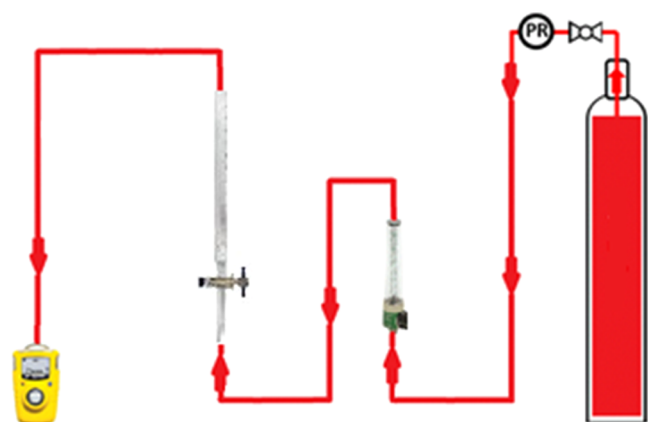


Figure 2. Setup of the H₂S-scavenging test.

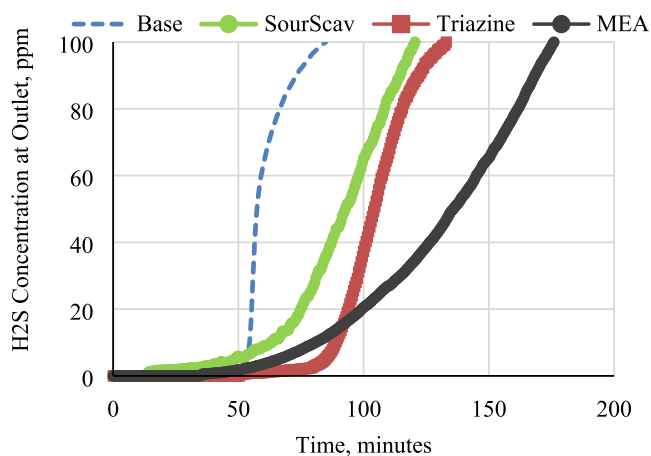


Figure 3. Results of hydrogen sulfide sorption test.

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.

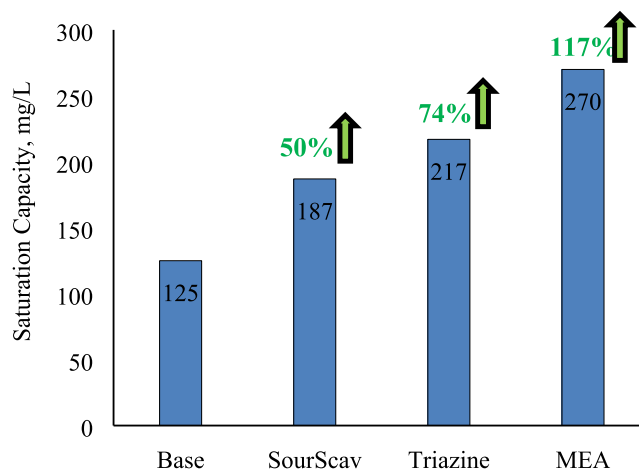
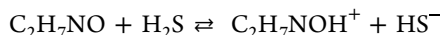


Figure 4. Resulted saturation capacities.

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^{19,72–75}



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).

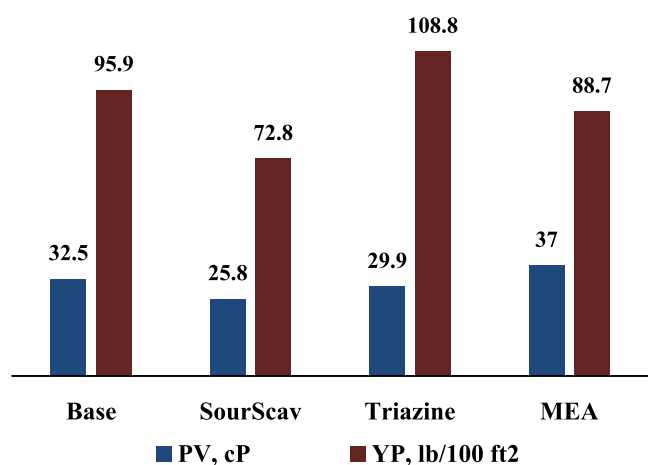
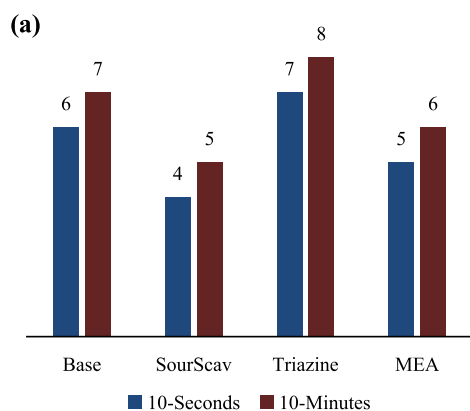


Figure 5. Resulted plastic viscosity and yield point.

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.⁶⁸ 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).

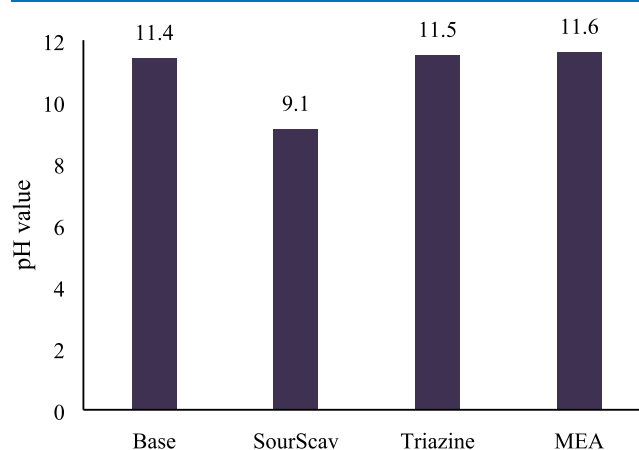


Figure 7. Results of pH measurement.

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.⁷⁶ 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

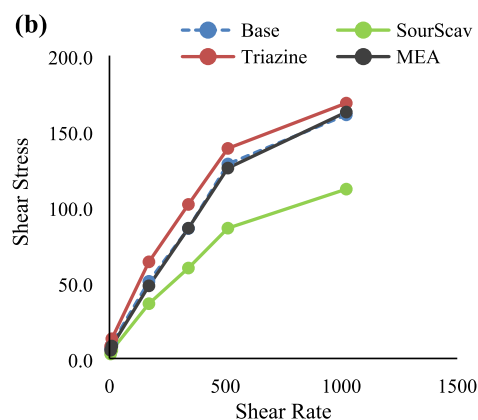


Figure 6. (a) Resulted gel strengths and (b) relationship of shear stress—shear rate.

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.^{72,75}

4.5. Filtration Test. The filtration test for the base mud revealed that the filtrated volume was 11.8 cm³ and the formulated filter cake had thickness and weight of 3 mm and 22 g, respectively. The addition of SourScav 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

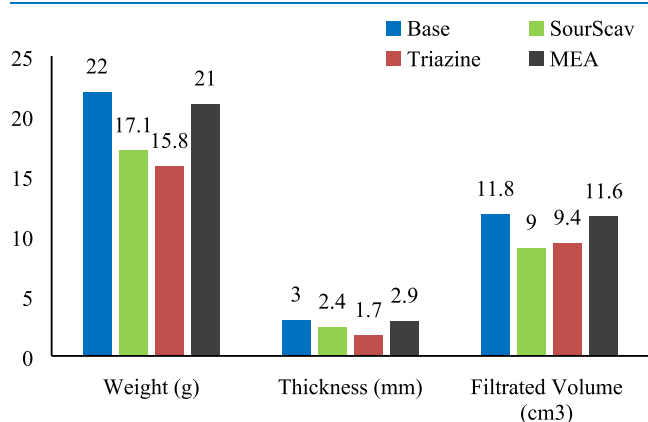


Figure 8. Results of the filtration test.

decrease in the filtrated volume to 11.6 cm³. 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 H₂S 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, SourScav, and triazine muds with the following conclusions:

- Using MEA considerably improved the H₂S-scavenging capacity by 117% relative to the base mud, while the commercial SourScav 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; SourScav and triazine also displayed noncorrosive behavior.

- Adding MEA to the base drilling mud slightly enhanced the filtration performance; however, SourScav and triazine provided preferable filtration characteristics.
- Further research and optimization of the mud formulation are required before field application trials.

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Author Contributions

S.E. and S.A.O. conceptualized the study. A.A. developed the methodology, performed the formal analysis and investigation, curated the data, wrote the original draft, and visualized the experiments. S.E. and S.A.O. gathered the resources. S.A.O. wrote the original draft and reviewed and edited it. S.E. supervised the experiment. All authors have read and agreed to the published version of the manuscript.

Notes

The authors declare no competing financial interest.

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NOMENCLATURE

API	American Petroleum Institute
C _{out}	H ₂ S concentration at the outlet gas stream, ppm
DEA	diethanolamine
HPHT	high pressure and high temperature
MDEA	methyl diethanolamine
MEA	monoethanolamine
NIOSH	National Institute for Occupational Safety and Health
OSHA	Occupational Safety and Health Administration
PV	plastic viscosity, cP
SRB	sulfate-reducing bacteria
t _s	saturation time, min
YP	yield point, lb/100 ft ²
ρ	H ₂ S density

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