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Development In The Scavenging Efficiency Of H_2S Scavengers In Oil And Gas Industry

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Abstract

Hydrogen sulfide H_2S is an acid gas present in oil and gas reservoirs. The presence of H_2S reduces the quality of natural gas. Hydrogen sulfide scavengers are widely used in the production and processing operations in the Oil and Gas Industry to maintain the integrity of the pipelines and workplace safety. Scavengers are divided into regenerative and non-regenerative. In degenerative scavengers, concentrated solutions of aqueous alkanolamines are the most employed for H_2S removal from natural gas. Non regenerative scavengers include triazines, amine based, aldehydes, metal chelates and others. Triazines represent the most widely used non regenerative scavengers due to their high efficiency, fast kinetics, and low toxicity. This paper includes the up to date development in the field of H_2S scavengers used to minimize or eliminate the various problems caused by hydrogen sulfide during oil and gas drilling operations. The advantages and disadvantages of such scavengers will be compared.

Keywords: Hydrogen sulfide, scavenger, development, natural gas, oil.

1. Introduction

Hydrogen sulfide H_2S is an acid gas present in oil and gas reservoirs. There are three sources of H_2S in the petroleum reservoirs. Sulfate reduction either by sulfate reducing bacteria or by a thermochemical process that involves sulfate reduction by hydrocarbons is the first source of H_2S. H_2S can also be formed in the reservoir by thermal cracking in which large organo-sulfur molecules are broken down into smaller molecules that later change into H_2S (Agbroko, Piler, & Benson, 2017). The presence of H_2S and CO_2 reduces the quality of more than 40% of natural gas reservoirs (Subramaniam et al., 2018). Thousands of parts per million (ppm) of H_2S can be found in produced gas and in some reservoirs, the H_2S level reaches 50% or more (G. Taylor, Smith-Gonzalez, Wylde, & Oliveira, 2019). During the production of crude oil, H_2S is released into pipelines and surface equipment, leading to several problems. H_2S is poisonous and can cause eye and throat irritation, paralysis, coma, and immediate collapse or death which creates huge safety and environmental concerns for the producers (E. Lim, Mbowe, Lee, & Davis, 2016). Furthermore, H_2S is highly corrosive due to its acidic nature as shown in equations 1 and 2.

$$H_2S + H_2O \rightleftharpoons H_3O^+ + HS^-;\ \mathrm{pKa} = 6.9 \quad (1)$$

$$HS^- + H_2O \rightleftharpoons H_3O^+ + S^{2-};\ \mathrm{pKa} = 13.9 \quad (2)$$

It can cause different kinds of corrosion, such as pitting and sulfide stress cracking (Saji, 2021). It can also contribute to the diffusion of elemental hydrogen into the surface of the pipelines causing hydrogen embrittlement. Iron sulfide which is a corrosion byproduct can deposit onto the surface of the pipeline causing under-deposit corrosion and potentially plugging the production equipment (C. Menendez, Jovancicevic, Zhu, Morton, & Stegmann, 2011). The deposits can interfere with the function of the system or equipment decreasing the efficiency of the production process (Saji, 2021). The presence of H_2S significantly increases the operation costs for sour oil and gas production. The increased costs are due to the need for metallurgical upgrades as the surface equipment should meet National Association of Corrosion Engineers (NACE International) standards for sour service. Gas sweetening equipment, manpower for monitoring, chemicals cost, and possible fines for oil not meeting the standards also add up to the oil and gas production costs (S Lehrer et al., 2015). Removing H_2S downhole and preventing it from reaching the surface can help alleviate these issues. Lowering the concentration of hydrogen sulfide can allow the use of less-expensive, corrosion-resistant alloys. Due to its toxic nature and the effects it has on the production equipment and on the environment, H_2S concentration in the natural gas should be limited to below 4 ppm as per the sales gas specifications (Scott Lehrer, Jani, Ramachandran, & Liu, 2021; Rincón, Jiménez-Junca, & Duarte, 2016).

In this review, developments, advantages, and disadvantages of H_2S scavengers utilized in the production of oil and gas are presented. The presented scavengers include triazine based, amine based, metal based, and others.

2. H_2S scavengers

The typical method used for H_2S removal from oil and gas industries involves the use of sulfide scavengers (Groysman, 2017; Scott Lehrer et al., 2021; Yang et al., 2014). The sulfide or H_2S scavengers are chemicals that can react with different sulfide compounds forming inactive or less toxic species (Saji, 2021). Scavengers can be solids that act as adsorbents for the H_2S or liquids that absorb H_2S and other sulfide compounds (Agbroko et al., 2017; S. K. Lim et al., 2021; Perez-Pineiro, Cruz-Perez, Hoshowski, Zhang, & Hendry, 2018). A proper H_2S scavenger should have a rapid and complete reaction with H_2S at low treatment levels and in a wide variety of pressure and temperature conditions. The reaction should not produce byproducts that may hold environmental concerns (Groysman, 2017; Jenkins et al., 2021; Jones, Sorrells, & Stark, 2017). It shouldn't cause fouling or corrosion and it shouldn't require major alterations to the production facility (Agbroko et al., 2017). H_2S scavengers can be classified into regenerative and non-regenerative scavengers. Regenerative scavengers are used in large facilities for the removal of high concentrations of H_2S. They involve the use of a fixed bed filled with absorbent material (Agbroko et al., 2017; Groysman, 2017). Regenerative scavengers are mostly used in gas streams in a gas sweetening process. The process involves an amine contractor tower that contains an alkanolamine as the scavenging chemical mixed with an activator. Such alkanolamines include monoethanolamine (MEA) and diethanolamine (DEA). The activators can be polyalkyleneamines, polyamines, alkoxypropylamines (Agbroko et al., 2017; Saji, 2021). Alkanolamines undergo reversible reaction with acid gases and can therefore be regenerated in a cyclic process by applying heat. Non-regenerative scavengers are chemicals that react irreversibly and completely with H_2S yielding non-lethal and non-corrosive compounds. They are water-soluble and are used for the production of oil and gas with low concentrations of H_2S. Such non-regenerative scavengers include triazines, aldehydes, and metal-based scavengers. Lots of research has been done to improve the overall performance of H_2S scavengers through increasing the scavenging capacity, reducing nitrogen contamination, reducing the formation of scale, preventing deposition of by-products, addressing existing environmental, health and safety concerns (Jenkins et al., 2021; Perez-Pineiro et al., 2018). New H_2S scavengers should have fast kinetic reaction and consumption rates. They should be compatible with the production system and should cause minimal precipitation of by-products. It should also be scalable and economically favorable (Hoshowski et al., 2021; Jenkins et al., 2021).

3. Research developments on H_2S scavengers

3.1. Triazine

Triazines represent the most widely used scavengers today (S Lehrer et al., 2015; G. N. Taylor, 2014). Formed by the reaction of formaldehydes with amines, Hexahydro-1,3,5-tris(2-hydroxyethyl)-s-triazine (MEA-triazine) is the most utilized hydrogen sulfide scavenger accounting for at least 80% of the available oilfield market (Benhabib, Kleinman, & Peterman, 2021; G. Taylor et al., 2019). They are considered noncarcinogenic (Saji, 2021). Triazines are efficient, inexpensive, and react quickly with H_2S (S Lehrer et al., 2015; G. N. Taylor, 2014). They can be altered to fit for the application both down-stream and up-stream of the wellbore (Madsen, Jensen, & Søgaard, 2014). The byproducts from the reaction of Triazine with H_2S are relatively non-toxic and biodegradable (Saji, 2021). However, triazines can cause plenty of undesirable effects and issues during the production and in refinery systems. For instance, some of the reaction byproducts such as amorphous dithiazine can cause solid deposits and fouling (Figures 1 and 2). Triazines along with their reaction byproducts can increase the pH of the water due to the presence of amine groups in their structure, causing scaling, deposition of reactive polymeric substances, and increased concentration of ethanolamine (G. Taylor et al., 2019). Furthermore, at low pH triazines undergo hydrolysis before reacting with H_2S which significantly reduces their scavenging efficiency (Saji, 2021). They have relatively slow reaction rates when used in direct injection applications, which reduces their effectiveness in systems with short residence times, resulting in the amount required being substantially more than the theoretical efficiency would imply (S Lehrer et al., 2015). Due to the low thermal stability of triazines and to their potential of forming scale, they cannot be used for downhole application (S Lehrer et al., 2015). Triazines can cause corrosion in refinery processes as they can form amine hydrochloride salts (Ramachandran, Lehrer, Chakraborty, & Jani, 2018).

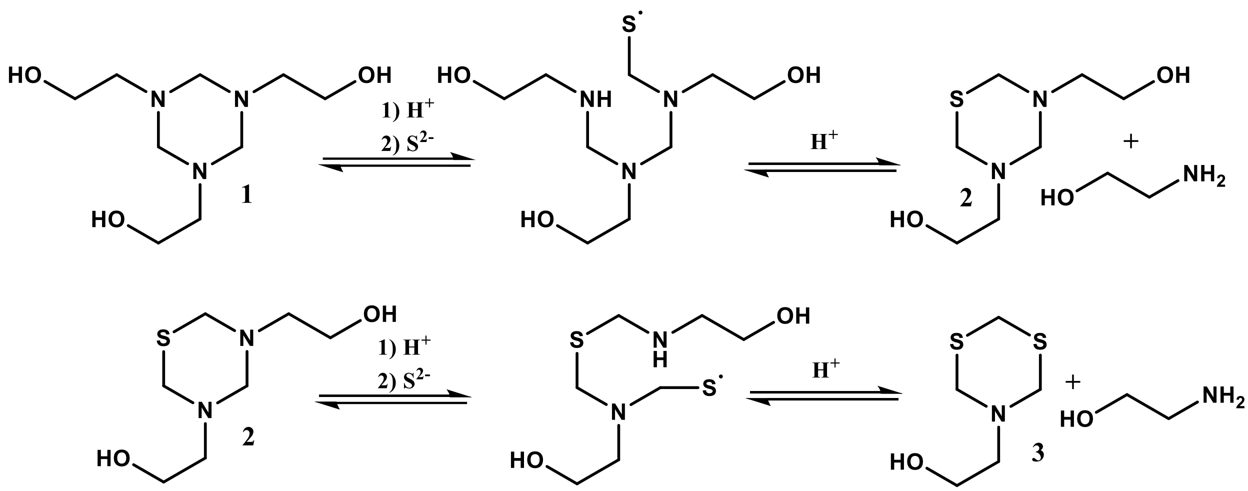

Figure 1. Formation of thiadiazine/dithiazine from the interaction between hexahydro-1,3,5,-tris(hydroxyethyl)-s-triazine and H2S.

Figure 2. Drain pipeline plugged with amorphous dithiazine.

As a result of the triazines limitations and disadvantages, plenty of research has been done to modify, replace or increase the efficiency of triazines. New scavengers known as Non-Triazine Based Scavengers (NTBS) have been developed. Taylor et al have developed a family of NTBS which can work as multicomponent systems that have a designated function each. The exact formula for NTSB scavengers was kept proprietary however, it was mentioned that the formulation contained a base scavenger, a synergistic component, a solid suppressant designed to prevent the formation of polymeric solids, and a suitable co-solvent. The active component is based on a concealed version of a small molecule scavenger (SMS) that remains in low concentrations and is released encountering H_2S in its operational environment. The release of the SMS can be significantly improved with the use of a suitable catalyst or synergist and the developed NBTS showed significantly higher efficiency than triazine (G. Taylor et al., 2019). Zaid et al patented a triazine chemical package that is water-dispersible and oil-soluble. The package contains triazine, glycol ether compounds to reduce corrosion and fouling, and optionally an alcohol (Ramachandran et al., 2018). Menendez et al patented a synergistic combination consisting of triazine and dialdehyde (e.g. glyoxal) (C. M. Menendez, Jovancicevic, & Ramachandran, 2016). The synergetic combination showed higher H_2S and mercaptans removal than dialdehyde alone or triazine alone as shown in Figure 3.

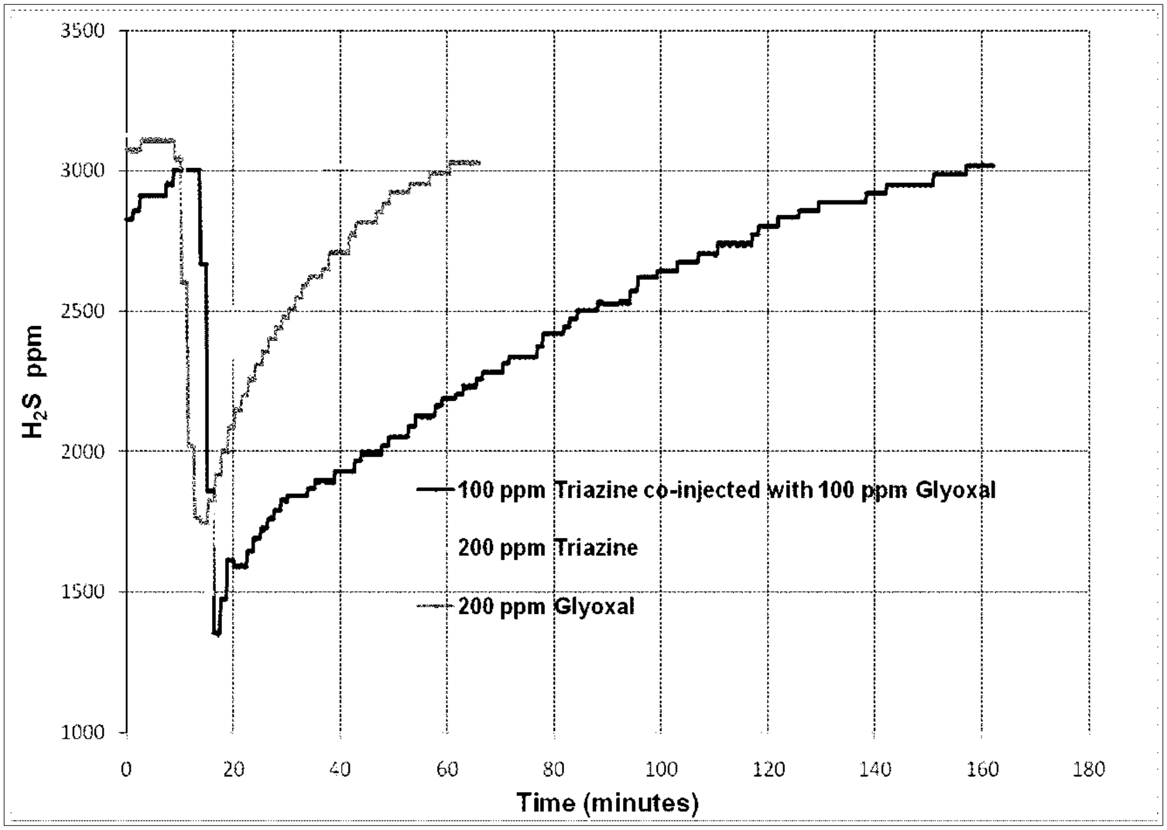

Figure 3. H_2S removal efficiency for 100 ppm glyoxal, 100 ppm triazine, and 1:1 mix of triazine and glyoxal.

Subramaniam et al developed a simulation program that computes the quantity of triazine required per pound of H_2S removal at various depths of the gas well, allowing for the determination of the optimum depth to achieve maximum H_2S removal with the least amount of triazine (Subramaniam et al., 2018).

3.2. Metal-based scavengers

Metal-based scavengers are primarily used in bitumen at T > 150°C and can easily blend into hydrocarbon streams. However, If not well monitored, they can cause fouling along with their reaction byproducts (Kenreck, 2014). Metal-based scavengers include metal salts, metal oxides, metal carboxylates, and chelates. Metal chelates with high valence metal ions have been employed in drilling fluids, contaminated water, and oil streams to treat the issues associated with the presence of H_2S (Agbroko et al., 2017). Chelates include carboxylate groups. Common metal ions include zinc(II) and iron(III). Absolute conversions of H_2S have been achieved by polycarboxylate-chelated iron process (Agbroko et al., 2017). Several patents were registered on metal-organic compositions/compounds. Lehrer et al developed a new organic acid metal complex based H_2S scavenger (OAC). The performance of the scavenger represented in two parameters was tested. The first parameter is the maximum ppm H_2S reduction correlating to the kinetics of the reaction with H_2S. The second parameter is the specific consumption of H_2S scavenger, expressed as gallons of scavenger per pound of H_2S (gal/lb). The testing was done at 167 F which is the temperature representing downhole injections. The performance of 200 ppm of OAC, 200 ppm of an oil-soluble amine aldehyde scavenger (AAS), 400 ppm of glyoxal, and 400 ppm of triazine was compared with each other (Figure 4).

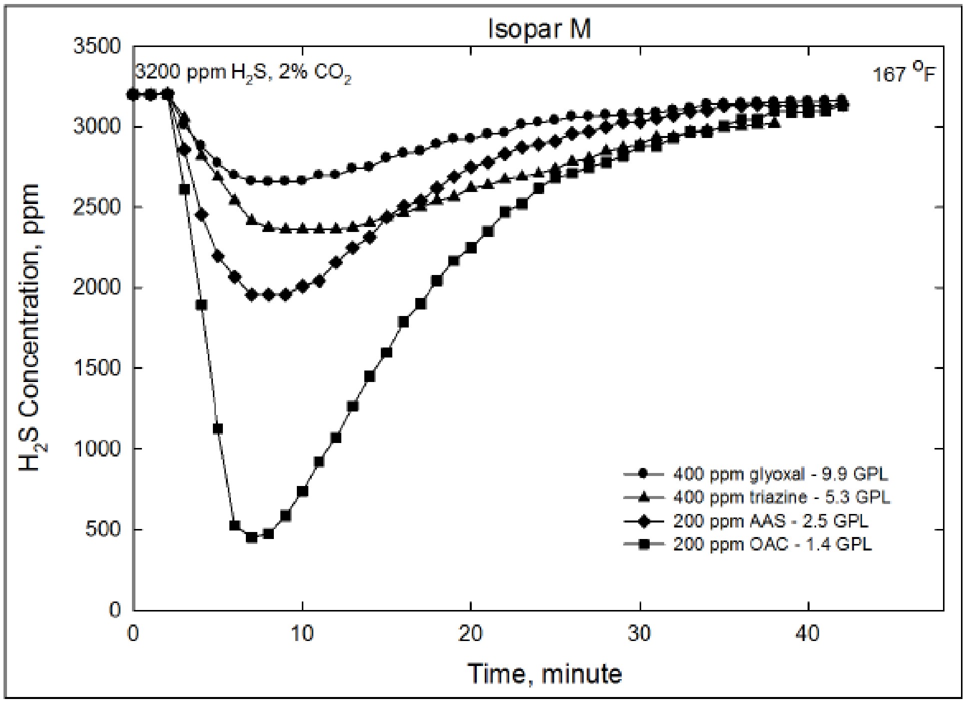

Figure 4. A comparison between H_2S scavengers.

Glyoxal and Triazine were tested using double the concentration due to their low performance at such conditions. As interpreted from Figure 4, the OAC showed the highest efficiency. The OAC has been found to rapidly react with H_2S effectively removing H_2S in high-brine systems and at high temperatures. The major advantage of the OAC scavenger is that it does not cause the formation of scale. Furthermore, the scavenger was found to be compatible with all metallurgies and most elastomers and plastics. In a case study, an oil well in South Texas produced 160 ppm of H_2S as shown in Figure 5. Despite using triazine at a high loading rate, the producer couldn't reduce H_2S to the specification limit of 10 ppm. Alternatively, OAC was injected into the wellhead and evaluated. OAC was able to reduce H_2S to below 10 ppm within 40 minutes (S Lehrer et al., 2015).

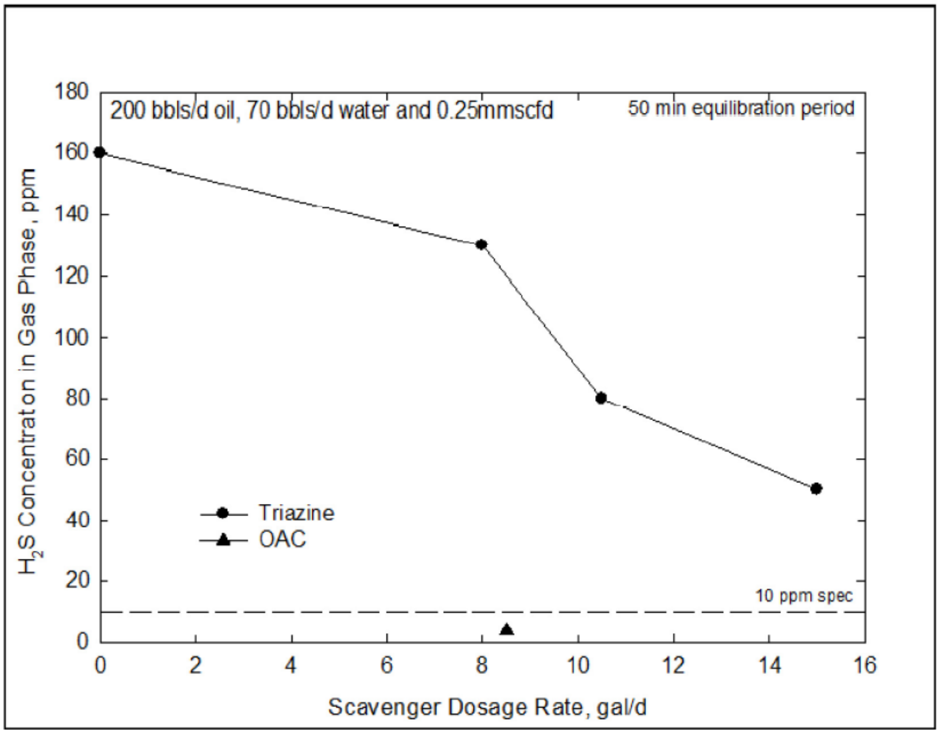

Figure 5. The application of OAC in South Texas oil-producing well.

Subramaniyam et al patented an H_2S scavenging composition in which an amine compound such as aliphatic tertiary amine is mixed with a Cu compound to act as an activator improving the scavenging efficiency (Subramaniam et al., 2018). Elkatatny et al tested the efficiency of copper nitrate as a non-corrosive H_2S scavenger in drilling fluids. H_2S reacts with copper nitrate according to equation 3.

$$H_2S + Cu(NO_3)_2 \rightarrow CuS + 2HNO_3 \quad (3)$$

The copper sulfide (CuS) is later removed from the fluid by filtration. Copper nitrate is then regenerated by reacting copper sulfide with hot nitric acid (HNO_3) forming elemental sulfur as a white precipitate. When compared with triazine, copper nitrate exhibited double the adsorption capacity. The addition of $Cu(NO_3)_2$ improved the drilling fluid properties; reducing the thickness of the filter cake and increasing the yield point/plastic viscosity which can improve the carrying capacity of the drilled cuttings from the downhole to the surface and the process of the hole cleaning. Recovery of copper and the removal of elemental sulfur can easily be done at the rig site at the surface conditions (Elkatatny, Basfer, Shawabkeh, Bahgat, & Mahmoud, 2019). Huang et al proposed Ferrocene as a vapor phase transported reagent for the removal of H_2S in situ. Ferrocene reacts completely and rapidly with H_2S forming iron sulfides, cyclopentadiene, and hydrogen. The reaction products are inert; however, iron sulfide may deposit and reduce permeability. Ferrocene has been shown to be an effective H_2S removal reagent, as no H_2S was detected from samples with added ferrocene (Huang, Zhang, & Han, 2021). Lehrer et al developed a scavenging mixture made of a transition metal salt such as zinc or iron carboxylates and at least one water-soluble aldehyde or water-soluble aldehyde precursor such as ethylene glycol hemiformal. The combination resulted in a significant increase in the reaction rate, overall scavenging efficiency, and capacity (S. E. Lehrer, Jovancicevic, & Ramachandran, 2017).

3.3. Amine based scavengers

Alkanolamines such as MEA, DEA, and MDEA represent the most commonly utilized scavengers in gas sweetening due to their safety and low cost (Shoukat, Pinto, & Knuutila, 2019). Just like triazines, the use of amines has limitations and can cause several issues during production. For instance, amine-based compounds can raise the pH of the medium and increases the potential for scale formation. They may break down at high temperatures (>150°C) or when used for long circulation. The products formed from the reaction of amine compounds with H_2S can be corrosive. In addition, heat stable amine salts can react with other chemicals and contaminants in the stream causing accumulation in the amine solution, resulting in corrosion with time (Groysman, 2017). The reaction of Amine compounds with H_2S can be reversed at high temperatures and low pH (Saji, 2021) as shown in equation 4.

$$\mathrm{H_2S} + \mathrm{R_3N} \rightleftharpoons \mathrm{R_3NH^+} + \mathrm{HS^-} \quad (4)$$

Due to the adverse effects of amines, several researches have been done for developing synergetic compositions to enhance their performance and avoid the issues associated with their use. Clark et al presented a scavenging mixture that includes an amine-containing compound and a hemiacetal compound (Clark JC & RC, 2019). Primary and/or secondary amines produced from the scavenging reaction react with the oxymethylene compounds in situ, as a result the amount of amines in the production fluid is reduced. A scavenger that is soluble in oil made by Zaboikina et al made via the reaction of a gaseous secondary amine such as dimethylamine and an aromatic aldehyde has been found to be of high efficiency (Zaboikina, Klyatskii, Maksimov, Tsvetkov, & Yudin, 2014). Kaplan et al presented a scavenger composition along with a method for their removal from the hydrocarbon fluid. The mixture comprises an amine-aldehyde reaction product and one oxymethylene compound (Kaplan G, 2018).

3.4. Others

Other H_2S scavengers include aldehyde-based scavengers preferably used in systems with acidic pH. Most commonly used aldehydes are formaldehyde, acrolein, glyoxal, and glutaraldehyde. Formaldehyde and acrolein are toxic. The reaction of Formaldehyde with H_2S forms solid products. Subramaniyam et al patented a glyoxylic acid-polyethylene glycol scavenging composition that forms water-soluble products, preventing fouling and decomposition issues (M., 2014).

Membranes have been used in a vast number of applications during the last few decades due to their easy operation and design flexibility. They control the movement of species acting as a selective boundary. A hollow fiber membrane contactor (HFMC) is a new membrane method for the separation of gases. In HFMC, the fluid containing an absorbent and the gas to be treated are made to flow in opposite sides of the membrane contactor (Figure 6). They come in contact at the surface of the membrane within the membrane pores. The hydrophobicity of the membrane plays an important role in the efficiency of the process due to its contact with the fluid. Hydrophobic materials used for membranes include Polypropylene (PP), Polyethylene (PE), Polytetrafluoroethylene (PTFE), and Polyvinylidene fluoride (PVDF). PVDF and PTFE membranes resemble more hydrophobic, permeable, and thermally stable membranes in comparison with PP and PE membranes. However, due to the complicated manufacturing of PTFE, it has significant production costs, which made PVDF membranes more industrially accepted.

Abdolahi-Mansoorkhani et al created a novel PVDF membrane structure using $CaCO_3$ nanoparticles. The nanoparticles were used to increase the efficiency of removal of CO_2 and H_2S from natural gas by increasing the porosity of the membrane which increases the contact area for the reaction. The membrane consisting of 20% $CaCO_3$ was found to have the highest efficiency. The tubes contained MEA as the absorbent fluid (Abdolahi-Mansoorkhani & Seddighi, 2019).

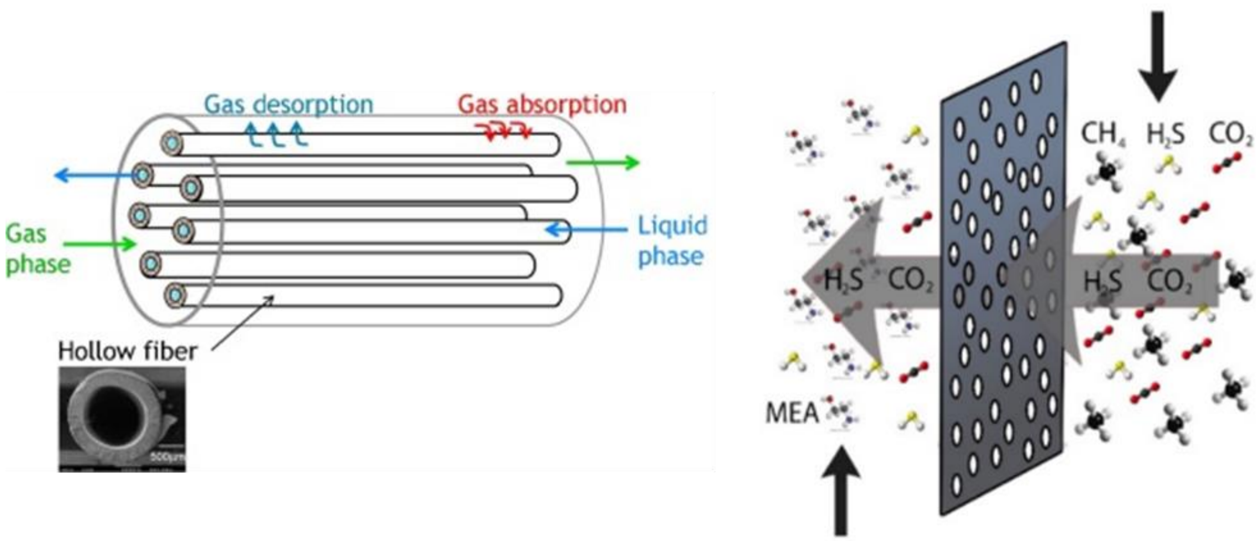

Figure 6. Geometrical configuration and schematic separation of Hollow Fiber Membrane Contractor's.

Nassar et al made an environmentally friendly nanocomposite compromised of co-polymers based on acrylamide (AM) and vinyl acetate (VA) and CdO nanoparticle. Three different types of (AMVA) copolymers were synthesized by free radical polymerization method each with a different molar ratio of AM to VA (Nassar, El-Din, Morsi, Abd El-Azeim, & Hashem, 2016).

4. Conclusion

H_2S scavengers are essential for the removal of H_2S from produced water, gas, crude oil, and refinery products to maintain the integrity of the pipelines and a safe workplace. Scavengers can be regenerative or non-regenerative. Concentrated solutions of aqueous alkanol amines are the most commonly employed for regenerative sweetening of produced gas. Non regenerative scavengers include triazines, amine based, aldehydes, metal chelates, and others. Triazines represent the most widely used non regenerative scavenger due to their high efficiency, fast kinetics, and low toxicity. Each scavenger kind has limitations and can cause issues such as scaling or corrosion in the production system. Developments in the field of H_2S scavengers were mostly registered patents for different scavenging packages and compositions. Recent research also included novel adsorbents and nano materials to increase efficiency and avoid issues associated with traditional H_2S scavengers.

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