

Factors Effecting Generation of H₂S During Storage in Marine Furnace Oil**Abhinav Bhandari***, Pankaj Pathak, Himanshu Gupta, Laxmi Bhandari, D Himanshu Athey, Amit Mishra, A R Mishra, V Singhal, V Suresh and A K Tiwari

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Excessive levels of hydrogen sulfide in the vapor phase above residual fuel oils in storage tanks can result in health hazards, violation of local occupational health and safety regulations, and public complaint. An additional concern is corrosion that can be caused by the presence of H₂S during refining or other activities. Control measures to maintain safe levels of H₂S require a precise method for the measurement of potentially hazardous levels of H₂S in fuel oils. Hydrogen sulfide (H₂S) is a very dangerous, toxic, explosive and flammable, colourless, and transparent gas which can be found in crude oil and can be formed during the manufacture of the fuel at the refinery and can be released during handling, storage, and distribution. At very low concentrations, the gas has the characteristic smell of rotten eggs. However, at higher concentrations, it causes a loss of smell, headaches, and dizziness, and at very high concentrations, it causes instantaneous death. It is strongly recommended that personnel involved in the testing for hydrogen sulfide are aware of the hazards of vapor-phase H₂S and have in place appropriate processes and procedures to manage the risk of exposure.

1. Introduction

Furnace oil, also known as heavy fuel oil or residual fuel oil, is a complex mixture of hydrocarbons and various impurities or contaminations, including sulfur compounds. Essentially these impurities are removed or limited during the refining processes, upto minimum specification requirements. However, the sulfur species present in the FO reacts due to various factors as detailed below, to generate hydrogen sulfide during storage and transportation. The generation of H₂S in furnace oil indeed varies depending on the quantity & type of sulphur species present, side reactions of sulphur with various fuel components under high temperature and pressure conditions, type of crude oil being processed, etc. The presence and types of sulfur species in petroleum fuels significantly influence both the environmental impact, technical handling of these fuels and ensuring compliance with regulatory standards. The desirable limit for hydrogen sulfide (H₂S) in marine furnace oil is generally considered to be 2 parts per million (ppm) or less. This limit is specified in international standards of ISO 8217:2017 standard, which is widely recognized in the maritime industry. Other organizations and classification societies, such as CIMAC, DNV GL, and ABS, also refer this ISO standard. MARPOL Annex VI indirectly specifies H₂S limits by referencing to ISO 8217 for fuel quality standards, which includes the H₂S limit. Measurement and testing of H₂S is typically carried out by IP 570 or ASTM D5705, with former being mentioned in

the ISO specification.

The 2 ppm (parts per million) limit for hydrogen sulfide (H₂S) in marine furnace oil is established for several critical reasons:

- Safety: H₂S is highly toxic and corrosive. Even small amounts can be dangerous to human health and equipment. At concentrations above 100 ppm, it can cause respiratory paralysis and death.
- Corrosion: H₂S can cause severe corrosion in storage tanks, pipelines, and engine components.
- Environmental concerns: When burned, H₂S produces sulfur dioxide (SO₂), which contributes to air pollution and acid rain.
- Quality indicator: The presence of H₂S can indicate poor refining processes or contamination of the fuel.
- It can reduce the overall quality and performance of the fuel.

1.1. If H₂S Levels Exceed 2 Ppm (Parts Per Million) In Marine Furnace Oil, Several Issues Can Arise

- Increased health risks for crew members, potentially leading to respiratory problems or even fatalities in severe cases. It's hazardous to crew health and safety.
- Accelerated corrosion of fuel systems, storage tanks, and engine components, leading to increased maintenance costs and potential equipment failures.
- Higher levels of SO₂ emissions when the fuel is burned, potentially violating environmental regulations, and contributing

to air pollution.

- Possible fuel rejection or non-compliance with international maritime fuel standards, leading to delays and additional costs for ship operators.
- Potential damage to catalysts or other emission control systems designed to reduce sulfur emissions from ships.
- Legal and financial liabilities for fuel suppliers and ship operators.
- Higher maintenance costs and potential equipment failures.

2. Causes of H₂S generation

2.1. Types Of Sulfur Species

Sulfur in Petroleum Fuels Are Categorized Based on Their Structure and Bonding with Hydrocarbons:

- Thiols (Mercaptans): Contain a sulfur atom bonded to a hydrogen atom and a carbon atom attached to aliphatic or aromatic hydrocarbons and are typically present in low concentrations
- Sulfides (Thioethers): Contain a sulfur atom bonded to two carbon atoms and found in both aliphatic and aromatic compounds.
- Disulfides: Two sulfur atoms bonded together Usually derived from the oxidation of sulfides or thiols. These compounds can form during refining processes and can be more stable than sulfides and thiols.
- Thiones (Sulfones): Contain a sulfur atom double-bonded to an oxygen atom and is generally present in heavier fractions of crude oil.
- Aromatic Sulfur Compounds: contains Thiophene and their derivatives such as, Benzothiophene, dibenzothiophene, and other polycyclic aromatic sulfur compounds. These compounds contain sulfur within aromatic rings. They are generally more stable and less volatile, contributing significantly to the sulfur content in heavier fuels.
- Organosulfur Compounds: This category includes a range of sulfur-containing compounds that can be structurally diverse, including cyclic compounds and those with complex ring structures through various types of bonding (covalent) with hydrocarbons.

2.2. Impact Of Crude Oil Types on H₂S Generation in Furnace Oil

The potential for H₂S generation is different among different crude types:

- Low Sulfur Crude: Low sulfur crude generally results in furnace oil with lower H₂S generation potential. Low sulfur crudes can contain refractory sulfur compounds that are difficult to remove and may contribute to H₂S formation under certain conditions. Fewer sulfur compounds are converted to H₂S.
- High Sulfur Crude: High sulfur crude, on the other hand, leads to furnace oil with a much higher potential for H₂S generation. This is not just due to the higher overall sulfur content, but also because of the diversity of sulfur compounds present (thiols, thiophenes, etc.). Some of these compounds are more prone to thermal decomposition and H₂S formation and thus has higher potential for H₂S formation.
- High TAN (Total Acid Number) Crude: While Total Acid Number (TAN) is not a direct measure of sulfur content, high TAN

crudes can indirectly influence H₂S generation. The naphthenic acids present in high TAN crudes can increase corrosion in refinery equipment. This corrosion can release metal ions that may catalyze the conversion of sulfur compounds to H₂S. Moreover, the refining processes used to handle high TAN crudes might alter the distribution and reactivity of sulfur compounds in the resulting furnace oil.

2.3. Refining Processes

The type of crude oil significantly impacts the refining processes, which in turn affects the characteristics of the resulting furnace oil:

- Low sulfur crude may require less intensive desulfurization, potentially preserving more of the original hydrocarbon structure.
- High sulfur crude necessitates more aggressive desulfurization, which can alter the overall composition of the furnace oil, potentially making it more prone to H₂S formation under certain conditions.

2.4. Thermal Stability

Furnace oil derived from low sulfur crude tends to have better thermal stability. This means it's less likely to break down and form H₂S when exposed to high temperatures during use. While furnace oil from high sulfur crude, even after desulfurization, may retain sulfur compounds that are more susceptible to thermal decomposition, leading to higher H₂S generation during use.

In practice, refineries often blend different crude types to achieve a balance between cost and desired product specifications. This blending can also affect the potential for H₂S generation in the final furnace oil product.

3. Reactions Involved in Generation of H₂S

H₂S in FO can be formed during long-term storage, especially in warm conditions or due to Blending or Mixing different fuel batches leading to unexpected H₂S formation. The exact composition and concentration of these species can vary depending on the source of the crude oil and the refining processes used.

H₂S (hydrogen sulfide) can form in furnace oil through various mechanisms, primarily under high temperature and pressure conditions. The main chemical reactions involved are:

- Thermal decomposition of thiol compounds: At high temperatures, some sulfur species can break down to form H₂S: $R-SH \rightarrow R-H + H_2S$ (where R represents a hydrocarbon group)
- Hydrodesulfurization reactions: In the presence of hydrogen (which can be generated from hydrocarbons at high temperatures), sulfur compounds can be converted to H₂S: $R-S-R' + 2H_2 \rightarrow 2R-H + H_2S$
- Reduction of sulfoxides: Sulfoxides can be reduced to form H₂S under certain conditions. $R-SO-R' + 2H_2 \rightarrow R-H + R'-H + H_2S$
- Hydrogenolysis of Thiophenes. $C_4H_4S + 4H_2 \rightarrow C_4H_{10} + H_2S$

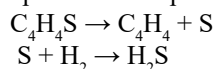
These reactions are simplified representations of complex processes. Actual mechanisms may involve multiple steps and intermediates, which is mentioned below: To elaborate on some key points from the artifact

3.1. Complex H₂S Formation Processes in Furnace Oil

- Thermal Cracking and Rearrangement:

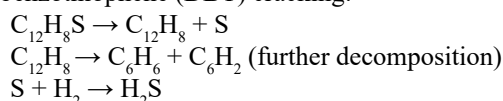
This process often starts with the breaking of carbon-sulfur bonds in larger molecules. The resulting fragments can then undergo rearrangement and hydrogen transfer reactions. For example, a complex thiophene derivative might break down into simpler thiophenes or even acyclic sulfur compounds, which are more reactive and prone to forming H₂S.

- Thiophene Decomposition:



The breakdown of larger sulfur-containing molecules like dibenzothiophene (DBT) often occurs through multiple steps. The initial C-S bond cleavage is followed by further decomposition of the resulting hydrocarbon fragments.

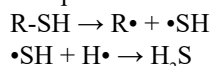
- Dibenzothiophene (DBT) cracking:



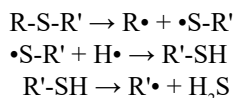
3.2. Free Radical Mechanisms

These are particularly important at high temperatures. The process might begin with the homolytic cleavage of a C-S bond, forming sulfur-centered radicals. These radicals can then abstract hydrogen from hydrocarbons or react with molecular hydrogen to form H₂S. The formation of sulfur-centered radicals can initiate chain reactions, leading to the formation of H₂S and various hydrocarbon products.

- Thiol decomposition:



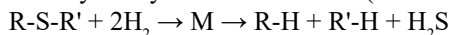
- Sulfide radical formation and propagation:



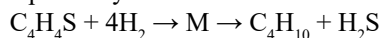
3.3 Catalytic Processes

Metals present in the furnace oil, either naturally occurring or from equipment corrosion, can catalyze desulfurization reactions. For instance, nickel and vanadium, common in heavy oils, can facilitate hydrodesulfurization reactions.

- Metal-catalyzed hydrodesulfurization (M = metal catalyst):



- Thiophene hydrodesulfurization:

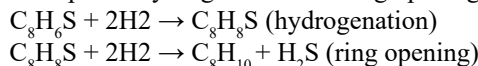


3.4. Influence of Other Compounds

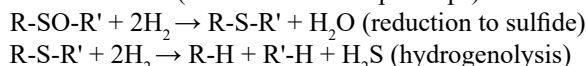
The complex hydrocarbon matrix of furnace oil plays a significant role. Aromatic compounds can stabilize certain sulfur species, making them more resistant to H₂S formation. Conversely, the

presence of hydrogen donors (like naphthenic compounds) can promote hydrodesulfurization reactions. The hydrogenation and ring-opening of benzothiophene illustrate how some sulfur compounds may need to go through several transformations before yielding H₂S.

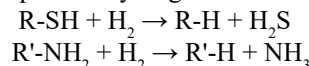
- Benzothiophene hydrogenation and ring opening:



- Sulfoxide reduction (can occur in multiple steps):

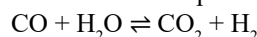


- Competitive hydrogenation with nitrogen compounds:



3.5. Temperature and Hydrogen Availability

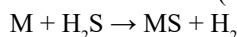
This is crucial for many H₂S formation pathways. In furnace oil at high temperatures, hydrogen can be generated through dehydrogenation of hydrocarbons or via the water-gas shift reaction if water is present.



3.6. Pressure Effects

Higher pressures can suppress some decomposition reactions that lead to H₂S formation. However, they can also promote hydrogenation reactions if sufficient hydrogen is available. The interplay of these factors makes H₂S formation in furnace oil a complex process that can vary significantly depending on the specific conditions and composition of the oil. This complexity is why precise prediction and control of H₂S formation often requires detailed analysis and modeling of the specific furnace oil in question.

- Metal sulfide formation (M = metal, such as Ni or V):



It's important to note that these reactions often occur simultaneously and can influence each other. The predominance of certain pathways depends on factors like temperature, pressure, and the specific composition of the furnace oil.

Note: R and R' represent hydrocarbon groups. These reactions are simplified and actual mechanisms may involve multiple intermediates.

4. Preventing or removing H₂S generation

H₂S in furnace oil is a critical concern for both environmental and operational reasons. There are several approaches to address this issue, ranging from preventive measures to removal techniques. H₂S Prevention and Removal in Furnace Oil are:

4.1. Prevention Methods

- Hydrodesulfurization (HDS) is the most widely used method in refineries. It's effective but can be energy-intensive and requires significant hydrogen input. The choice of catalyst and operating conditions can be tailored to the specific sulfur compounds present

in the furnace oil. Catalytic reaction of sulfur compounds with hydrogen is carried out which Converts sulfur to H_2S , which is then removed.

- Oxidative Desulfurization (ODS) is gaining attention as a complementary or alternative method to HDS, especially for treating oils with refractory sulfur compounds that are resistant to HDS. Oxidation of sulfur compounds followed by extraction or adsorption performed under mild conditions.
- Biodesulfurization is an emerging technology that shows promise for its selectivity and mild operating conditions, but it's not yet widely implemented on an industrial scale. Use of microorganisms to selectively remove sulfur which operates under mild conditions.

4.2. Removal Techniques

- Amine treatment is very common for H_2S removal from gas streams. In the context of furnace oil, it might be applied to treat vapors or associated gas streams. Absorption of H_2S into amine solutions is Heated to release H_2S , which is then typically converted to elemental sulfur.
- Adsorption methods can be effective for final polishing to achieve very low sulfur levels. The choice of adsorbent depends on the specific sulfur compounds present and other contaminants in the oil. Activated carbon, zeolites, metal oxides Physical or chemical adsorption of H_2S .
- Membrane separation is an area of active research, with potential for energy efficient H_2S removal, especially for gas streams associated with furnace oil processing. Selective permeation of H_2S through membranes, Low energy consumption, continuous operation.
- The Claus process is typically used for large-scale H_2S removal and sulfur recovery in refineries. It's more relevant to processing the H_2S removed from furnace oil rather than direct treatment of the oil itself. Partial oxidation of H_2S to elemental sulfur and water Typically used for large-scale H_2S removal.
- Liquid redox processes can be effective for moderate amounts of H_2S and are often used in smaller-scale operations or as a complement to other technologies. Oxidation of H_2S to elemental sulfur using a redox catalyst LO-CAT, Sulferox processes.

Note: The effectiveness of these methods can vary depending on the specific composition of the furnace oil and the operating conditions. Often, a combination of techniques is employed for optimal results.

It's worth noting that as environmental regulations become more stringent, there's ongoing research into developing more efficient and cost-effective methods for sulfur removal. This includes exploring new catalysts, process intensification, and novel hybrid approaches combining different technologies.

5. Reasons for High H_2S Generation in High Sulfur Crude

5.1. Higher Initial Sulfur Content

The most straightforward reason for increased H_2S generation is simply the higher concentration of sulfur compounds. More sulfur means more potential for H_2S formation. In high sulfur crude, you might find sulfur content exceeding 2% by weight, compared to

less than 0.5% in low sulfur crude.

5.2. Diversity of Sulfur Compounds

High sulfur crude doesn't just have more sulfur; it has a wider variety of sulfur compounds. This includes less stable forms like mercaptans (thiols) and sulfides, which are more prone to decomposition and H_2S formation. It also contains higher concentrations of thiophenic compounds, which, while more stable, can still contribute to H_2S generation under certain conditions.

5.3. Thermal Instability

The C-S bonds in many sulfur compounds are relatively weak compared to C-C or C-H bonds. In high sulfur crude, there are simply more of these bonds, increasing the likelihood of thermal decomposition during processing or use, especially at high temperatures.

5.4. Catalytic Effects

High sulfur crude often contains higher levels of metal contaminants like nickel and vanadium. These metals can act as catalysts for the conversion of organic sulfur compounds to H_2S . Additionally, the interaction between sulfur and these metals can create new catalytic sites that further promote H_2S formation.

5.5. Hydrogen Availability

Hydrodesulfurization (HDS), a common process for sulfur removal, requires hydrogen. With more sulfur to remove, there's a higher demand for hydrogen. If there's insufficient hydrogen available during processing, it can lead to incomplete desulfurization and the formation of reactive intermediates that are prone to H_2S generation.

5.6. Chemical Interactions

In high sulfur environments, there's an increased likelihood of sulfur-sulfur interactions. Some sulfur compounds can react with each other, leading to the formation of H_2S as a byproduct. There can also be synergistic effects where the presence of one type of sulfur compound catalyzes the breakdown of another.

5.7. Processing Challenges

To effectively process high sulfur crude, refineries often need to use more severe conditions (higher temperatures, pressures, etc.). While necessary for sulfur removal, these harsh conditions can also promote side reactions that lead to H_2S formation. The complexity of processing high sulfur crude can sometimes lead to incomplete desulfurization, leaving reactive sulfur species in the oil.

It's worth noting that the issue of H_2S generation from high sulfur crude is a significant challenge in the oil industry. It not only poses safety and environmental concerns but also impacts the economics of oil processing. As a result, there's ongoing research into more effective desulfurization technologies and methods to inhibit H_2S formation.

Understanding these mechanisms of H_2S generation is crucial for developing more effective strategies to mitigate the problem, both

during oil processing and in the use of oil products like furnace oil.

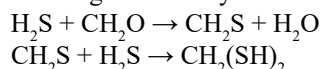
6. H₂S Scavengers: Types, Mechanisms, and Products

H₂S scavengers are an important tool in managing hydrogen sulfide in the oil and gas industry. Let's dive into the types of scavengers, their mechanisms, and some specific products.

6.1. Types of H₂S Scavengers

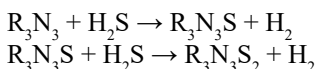
• Aldehyde-based Scavengers:

Aldehyde-based scavengers react with H₂S to form thioacetals or similar compounds (thiazolidines or dithiazines). This is an irreversible reaction, effectively removing H₂S from the system. Examples: Formaldehyde, Glyoxal, Acrolein. The reaction can be seen using formaldehyde as an example:



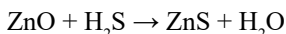
• Triazine-based Scavengers:

Triazine-based scavengers are popular due to their efficiency and relatively low cost. They react with H₂S to form stable, water-soluble products (dithiazines and other sulfur-containing compounds). Examples: 1,3,5-tri-(2-hydroxyethyl)-hexahydro-s-triazine. The reaction can be seen using Triazine as an example:



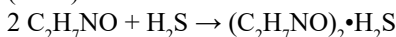
• Metal-based Scavengers:

Metal-based scavengers form insoluble metal sulfides, physically removing sulfur from the system. Examples: Zinc oxide, Iron oxide. The reaction can be seen using zinc oxide as an example:



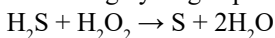
• Amine-based Scavengers:

Amine-based scavengers work through an acid-base reaction, forming amine sulfides. These are often used in gas treating processes. Examples: Monoethanolamine (MEA), Diethanolamine (DEA). The reaction can be seen using MEA as an example:



• Oxidizing Scavengers

Oxidizing scavengers convert H₂S to elemental sulfur or sulfate, depending on the oxidizer strength and conditions. Examples: Sodium hypochlorite, Hydrogen peroxide. The reaction can be seen using hydrogen peroxide as an example:



6.2. Commercial Products and Manufacturers

- Baker Hughes: SULFIX™ H₂S scavengers
- Schlumberger: H₂S SAFE™ scavenger
- Halliburton: COBRA™ H₂S Scavenger
- Innospec: OGS™ H₂S Scavengers
- 5. ChampionX (formerly Nalco Champion): EC6201A

The choice of scavenger depends on various factors, such as Temperature and pressure conditions, Phase of the system (gas, liquid, or multiphase), Presence of other contaminants,

Environmental regulations, Cost considerations, etc.

6.3. Efficiency And Reaction Kinetics: Different Scavengers Have Varying Reaction Rates and Efficiencies

- Triazine-based scavengers generally have fast reaction kinetics, making them suitable for many applications.
- Metal-based scavengers can have very high capacity but may react more slowly.
- Aldehyde-based scavengers are often chosen for their fast reaction rates but may have health and safety concerns.

6.4. Environmental and Safety Considerations

Some scavengers, particularly formaldehyde-based ones, have raised environmental and health concerns. This has led to increased interest in "green" scavengers that are more environmentally friendly.

6.5. Continuous vs. Batch Treatment

Scavengers can be applied in continuous treatment processes or as batch treatments, depending on the specific application and H₂S levels.

6.6. Monitoring and Control

Effective use of H₂S scavengers often requires careful monitoring of H₂S levels and scavenger dosage. Over-treatment can lead to unnecessary costs, while under-treatment may not adequately address the H₂S problem.

6.7. Emerging Technologies

Research is ongoing into new scavenger technologies, including:

- Nano-engineered materials for more efficient H₂S removal
- Biologically based scavengers using certain bacteria or enzymes.
- Combination products that address multiple issues (e.g., H₂S and CO₂) simultaneously

6.8 Product Selection

The specific products mentioned (SULFIX™, H₂S SAFE™, COBRA™, OGS™, EC6201A) are examples of commercially available scavengers. Each has its own formulation and may be more suitable for certain applications. It's important to consult with the manufacturers and conduct field trials to determine the most effective product for a specific situation.

7. Conclusion

In practice, managing H₂S often involves a combination of approaches, including scavengers, process optimization, and engineering controls. The choice of scavenger and overall H₂S management strategy should be tailored to the specific conditions of each application, considering factors such as H₂S concentration, system conditions, regulatory requirements, and economic constraints.

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