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Mercury Removal from Natural Gas in Egypt

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ABSTRACT

Worldwide natural gas is forecasted to be the fastest growing primary energy source. In Egypt, natural gas is recently playing a key role as one of the major energy sources. This is supported by adequate gas reserves, booming gas industry, and unique geographical location. Egypt's current proven gas reserves accounted for about 62 TCF, in addition to about 100 TCF as probable gas reserves. As a result, it was decided to enter the gas exporting market, where gas is transported through pipelines as in the Arab Gas pipelines project and as a liquid through the liquefied natural gas (LNG) projects in *Damietta*, and *Idku*.

With the start up of these currently implemented LNG projects that are dealing with the very low temperatures (down to -162 °C), the gas has to be subjected to a regular analysis in order to check the compliance with the required specifications.

Mercury is a trace component of all fossil fuels including natural gas, condensates, crude oil, coal, tar sands, and other bitumens. The use of fossil hydrocarbons as fuels provides the main opportunity for emissions of mercury they contain to the atmospheric environment; while other traces exist in production, transportation and processing systems.(1)

Worldwide mercury is present in natural gas wells to varying levels and its complete removal is needed especially for LNG projects. This is due to the following:

- Down stream aluminum heat exchangers that are subjected to corrosion in such LNG plants.
- Mercury vapor that causes sever corrosion to the plants' pipes and valves leading to the equipments' mechanical failure.
- Catalyst poising problems that are occurred due to the presence of mercury within the plants.
- Bad effect of mercury to the environment.

As very limited measures for assessing mercury presence in the Egyptian natural gas fields, a pilot research project is currently implementing aiming at achieving the following goals:

- Identifying different methods for determining mercury percentage in the gas.
- Identifying most appropriate techniques/approaches for mercury trapping and removal from the gas.
- Conducting field surveys for assessing mercury existence a long the gas chain in order to set the permissible limits of its existence.
- Establishing a database for mercury existence within different gas activities.

Introduction

The last five years have witnessed a significant increase of natural gas reserves in Egypt, where the proven reserves reached up to 62 TCF with about 100 TCF as probable reserves as reported by the end 2003. This was combined with large development and expansion in various gas activities all the way down the natural gas chain, including the establishment of two LNG plants at Idku and Damietta in addition to gas export through pipelines to Jordan by July 2003. This in turn necessitates the investment of large amounts of capital. Meanwhile, and although the analysis of natural gas covers the measurements of several gas constituents, however it does not include mercury.

Natural gas that is being produced from gas fields around the globe often contains mercury in addition to large number of impurities as CO_2, H_2S, RSH, sand, inert gases, and COS. Mercury is present in natural gas streams to varying levels. It is almost exists in its elemental form at low concentrations far below saturation suggesting that no liquid mercury phase exists in most gas reservoirs. In spite of that, its complete removal is needed in order to avoid its bad effects especially when dealing with operating conditions of very low temperatures.

Mercury Overview

Mercury is a naturally occurring metal which has several forms. Metallic mercury is a shiny, silver-white, odorless liquid. Mercury combined with carbon is called organic mercury; methyl mercury is a common example of organic mercury. Compounds which contain mercury in combination with non-carbon substances such as chlorine, oxygen, or sulfur are inorganic mercury compounds or "salts". These compounds are usually white powders or crystals

Metallic mercury is used to produce chlorine gas and caustic soda, and is also used in thermometers, dental fillings, and batteries. Mercury salts are sometimes used in skin lightening creams and as antiseptic creams.[1]

Mercury metal is a silver-gray liquid. Because of its unique properties as a liquid metal, metallic mercury is especially challenging to control. When exposed to the air or if spilled, mercury metal vaporizes into the air where it can be breathed into the lungs. The warmer the temperature, the more quickly the mercury gets into the air. A temperature increase from 64.4 F to 78.8 F doubles mercury's vapor pressure. Mercury can also be absorbed or injected through the skin, but is not usually harmful if it is eaten unless it becomes lodged in the digestive system. If spilled mercury is not cleaned up completely, it easily gets spread around.

Mercury forms droplets that can accumulate in the smallest spaces like cracks between floorboards. These droplets are very slippery and hard to remove from work surfaces or skin. If there is contamination of a worker with mercury, a micro environment of mercury vapor is created around that worker that can give exposure greater than that attributable to the general work environment. Mercury can be spread around work, car, and home from shoes, clothing, hair and other objects with tiny drops of mercury metal on them. [2]

In the environment, mercury levels are increased by certain human activities such as the burning of coal by power plants. Burning coal increases the amount of mercury in air, which eventually falls back to earth into bodies of water. Mercury in water accumulates in fish as the water passes over their gills. Common ways in which people are exposed to mercury include breathing contaminated air, eating contaminated fish, and through the use of mercury based amalgams (fillings) in dental treatments. Mercury can also enter the body through direct skin contact. Occasional exposures to mercury can also occur by contact with broken household items such as thermometers.

The effects of mercury on unborn children have been documented in accidental poisonings and scientific studies. During the 1950's, large amounts of organic mercury were dumped into the *Minamata Bay* in Japan, and fish containing high mercury levels were eaten by many pregnant women. Many of the children born to those women had severe nerve damage, which was later referred to as Fetal *Minamata* Disease. In Iraq, children born to mothers who ate grain contaminated with organic mercury may have learned to walk at a later age. In the Faroe Islands, where mercury exposure occurs primarily by eating whale meat, children born to mothers with higher body levels of mercury scored lower on brain function tests. However, no effects were seen in children of the Seychelles Islands, where residents are exposed to mercury by eating fish 12 times a week.

The Mercury Cycle

Trace amounts of elemental mercury occur naturally in the earth's crust. When rocks and soil deteriorate, this mercury is released. It is estimated that between one- and two-thirds of total mercury releases result from human activities, like mining and fossil fuel-burning.

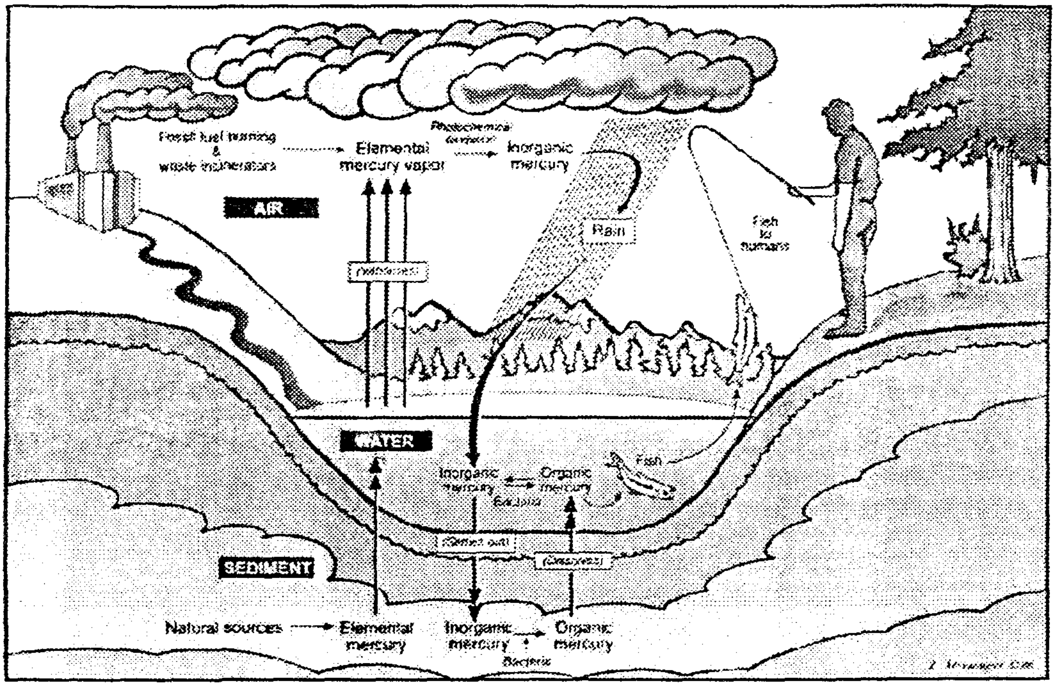
Of the mercury released by human activity about 80% is in its elemental form. Elemental releases result from mining, smelting, fossil fuel combustion, and solid waste incineration. About 15% is released from fertilizers, fungicides, and municipal solid waste. The remaining 5% is released from industrial wastewater.

Mercury is unique among metals in the fact that it is liquid at room temperature. Even at room temperature, some mercury evaporates. Mercury vapors can be carried long distances and deposited in rain or snow. Inorganic mercury can also enter water or soil from the weathering of rocks, wastewater containing mercury, and incineration of municipal waste containing mercury.

Microorganisms (including bacteria, phytoplankton, and fungi) convert inorganic mercury to methyl mercury. Released methyl mercury lingers in the water and soil. Once it has entered the food chain, methyl mercury bio-accumulates at progressive levels in the food chain. Fish that live a long time and attain large body sizes build up the highest concentrations of mercury within their bodies. Interestingly, even if grown in soils containing high levels of mercury, plants generally maintain very low mercury concentrations. [3]

Mercury moves through the environment as a result of both natural and human activities. The human activities that are chiefly responsible for causing mercury to enter the environment are burning mercury-containing fuels and wastes, and industrial manufacturing processes. Mercury emissions are transported through the air and deposited to water and land where humans and wildlife are exposed.

Methyl mercury is the most available form mercury is most toxic form to insects, fish, and humans. This form of mercury is easily taken up and accumulated in the tissues. Unlike many other fish contaminants mercury does not concentrate in the fat, but in the muscle tissue. Thus, there is no simple way to remove mercury-contaminated portions from fish that is to be eaten Mercury in the Environment. [4]



Sources and Cycling of Mercury to the Global Environment

The releases of mercury to the biosphere can be grouped in four categories, which are:

- Natural Sources: releases due to natural mobilisation of naturally occurring mercury from the Earth's crust, such as volcanic activity and weathering of rocks.

- Current anthropogenic (associated with human activity) releases from the mobilisation of mercury impurities in raw materials such as fossil fuels particularly coal, and treated/ recycled minerals.
- Re-mobilisation of historic anthropogenic mercury releases previously deposited in soils, sediments, water bodies, landfills and waste/tailings piles.
- Current anthropogenic releases resulting from mercury used intentionally in the different industrial activities such as;
    - ***Coal Fired Power Plants:*** the large volume of coal burned every year makes this the largest source in most countries.
    - ***Gold Refining:*** old methods of refining involved the use of very large amounts of liquid mercury in separation process.
    - ***Chlor-Alkali Plants:*** most of existing plants that are used as a part of pulp and paper mills used mercury as one of the electrodes in the electrolysis of brine to produce chlorine gas and sodium hydroxide.
    - ***Waste Incineration:*** it is a significant source, although emissions vary widely depending on the particular type of waste being burned.
    - ***Batteries Manufacturing:*** even the new mercury free batteries still contain considerable amounts of residual mercury
    - ***Fluorescent Lamps:*** the manufacture and disposal of these lamps have many risks due to its high mercury levels.
    - ***Electrical Industries:*** mercury is used in thermometers, lighting switches, relays, and other electrical equipments.

- *Mercury Mining:* mercury mining produces large amounts of the elemental form.
- *Petroleum Refining:* depending on the feed stock, high levels of mercury may be present in crude oil and natural gas as well as its finished products. [5]

Mercury Applications

Mercury Early Applications

Mercury was among the first metals known, and its compounds have been used throughout history. Archaeologists found mercury in an Egyptian tomb dating from 1500 BC. The Chinese may have been using mercury ores (mercury sulfide) as a red pigment for centuries before the birth of Christ. The Greeks knew mercury and used it as a medicine.

Mercury Modern Uses

The usefulness of mercury is limited by its poisonous nature. Mercury is used in electrical switches that are used in thermostats and some doze alarm type. It is highly suitable for use in thermometers because it doesn't moisten glass and its thermal expansion is uniform. Although many liquids could be used in pressure-measuring devices, mercury is used because its high density requires less space.

Mercury combines with numerous metals to form amalgam and is thus used to extract gold from rocks by amalgamating with the gold and then boiling off the mercury. The amalgam used in dental filling contains tin and silver amalgamated with mercury. Mercury vapor lamps are widely used where they are powerful and economical sources of ultraviolet and visible. As chemical compounds, it has many uses such as:

- in batteries (as a dioxide)
- biocides in paper industry and paints
- as antiseptics in pharmaceuticals
- laboratory analyses reactants
- catalysts
- pigments and dyes
- detergents [6]

Mercury Health Effects

Breathing mercury metal vapor over time affects the human brain, eyes, and kidneys. Inhaled mercury vapor may cause mood changes; inability to concentrate; memory loss; a fine shaking, tingling, or loss of feeling of the hand, tongue, or eyelid; discoloration of the cornea and lens of the eye; disturbances of vision; and kidney disease.

Children are more sensitive than other to mercury poisoning. With significant exposure, children can get "pink disease" with a rash over the body, chills, swelling and irritation of hands, feet, cheeks, and nose, light sensitivity, trouble sleeping, and heavy sweating.[2]

WORKPLACE EXPOSURE LIMITS

The following exposure limits are for Mercury vapor:

OSHA: The legal airborne permissible exposure limit (PEL) is 0.1 mg/m^3, not to be exceeded at any time.

NIOSH: The recommended airborne exposure limit is 0.05 mg/m^3 averaged over a 10-hour work shift and 0.1 mg/m^3, not to be exceeded during any 15 minute work period.

ACGIH: The recommended airborne exposure limit is 0.025 mg/m3 averaged over an 8-hour work shift [7].

* The above exposure limits are for air levels only. When skin contact also occurs, you may be overexposed, even though air levels are less than the limits listed above.

Mercury in Natural Gas

Mercury occurs naturally in trace quantities in air and natural gas. Mercury concentrations in natural gas are typically reported as microgram per "normal" cubic meter, where normal (N) indicates standard temperature and pressure. Mercury concentrations in natural gas vary from 0 to < 300 $\mu g/Nm^3$ with some of the highest concentrations occurring according to the gas geological origin. [8] Elemental mercury and organo-mercury compounds are present in natural gas in many regions of the world. The highest reported levels of mercury in gas are found in Southeast Asia, Eastern Europe, and North Africa [9] table (1) shows the reported mercury levels at different regions.

Table (6): Regional Average Level of Mercury in Natural Gas

Location	Elemental Mercury Concentration (Micrograms/m^3)
South America	69 – 119
Far East	58 – 193
North Africa	0.3 – 130
Gorningen	180
Middle East	1 – 9
Eastern US Pipeline	0.019 – 0.44
Midwest US Pipeline	0.001 – 0.10
North America	0.055 – 0.04

Problems Caused By Mercury in Natural Gas

It is reported that most if not all the mercury in natural gas is in the elemental form and that no natural gas processing plant problems are suspected to have been caused by organic or inorganic mercury compounds. This means that ***elemental mercury is the probable cause of mercury corrosion problems***.

Many reported cases all over the world (USA, Algeria, Holland, and Indonesia) describe huge failure resulting with great damage and environmental catastrophes as a consequence of uncontrolled presence of mercury in the gas plants especially in the cryogenic natural gas operations.

The implication of mercury effects in natural gas was not reported until 1973, when a catastrophic failure of aluminum heat exchangers occurred at Skikda liquefied natural gas plant in Algeria. Investigations determined that mercury corrosion caused the failure. After the Skikda failure, a study on Groningen fields in Holland revealed similar corrosion in the gas gathering system. CO_2 was initially thought to be the cause, but later investigation pointed to mercury with concentrations ranging from 0.001 to as high as 180 $\mu g/Nm^3$.

Although the concentration of mercury in natural gas may considered extremely low, it observes that its effect is cumulative as it amalgamates (forming alloys). Elemental mercury forms an amalgam with the surface layer of the metal it contacts. With aluminum, the corrosion problems occur when mercury comes into contact with an aluminum metal surface, where aluminum diffuses from the interface into the mercury droplet and it is rapidly converted to Al_2O_3 by reaction with air or water.

By this mechanism, metallic mercury actually bores into the aluminum leaving brittle layer of Al_2O_3. To initiate aluminum corrosion, the tightly adhering aluminum oxide layer on the surface of the aluminum must be removed. The mercury/aluminum amalgam process removes this oxide layer in the presence of a catalyst or an aqueous electrolyte.

The aqueous corrosion cell forms aluminum hydroxide and gaseous hydrogen through the following reactions:

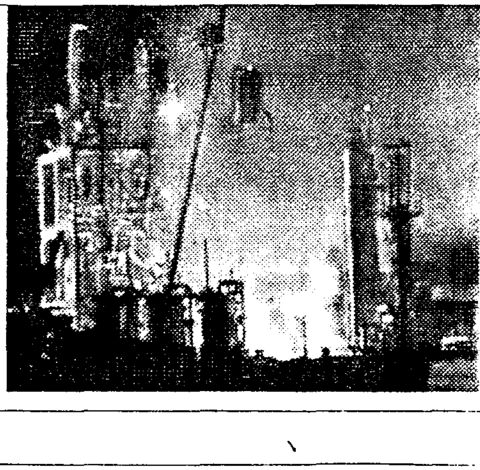
$$Al + Hg \longrightarrow Al\,Hg$$

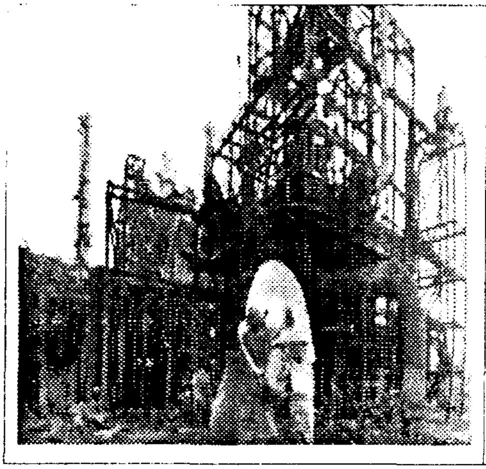
$$2AlHg + 6H_2O \longrightarrow 2Al\,(OH)_3 + 3\,H_2 + 2\,Hg$$

These reactions leave the previously amalgamated mercury free to form additional amalgam with base metal in a continuous process. Metal corrosion does not start at a significant rate if the equipment is maintained below the melting point of mercury (-39 °C). [9]

This problem is clearly shown in the natural gas liquefying processes (LNG), and greatly affects the processing equipments (vessels/pipes) specifically the aluminum heat exchangers used to liquefy the gas. As illustrated above, elemental mercury forms an alloy with aluminum (amalgam). This amalgam (which is much weaker than the metal itself) reacts with moisture to form a metal oxide plus free mercury, which can continue the corrosion process. Consequently, the mercury can damage the aluminum used in these exchangers and must be completely removed to no detectable levels in upstream equipment.

Out of the above described huge failures what had happed in Algeria (Skikda LNG complex) late January 2004.



The explosion was the worst accident at an LNG site in nearly 30 years ago, where it destroyed three out of six production trains. It was believed that a boiler at one of the gas trains was the origin of the blast, which was felt for miles and destroyed three of the refinery's LNG plants. The explosion badly caused damage at the nearby port where LNG tankers are loaded, and left at least 23 people dead, 74 injuries, and nine workers were still missing. Accordingly, all activities at the oil and gas refinery complex are shut down. Therefore, prevention of mercury entering into eco-technology-system is absolutely critical. One way for achieving this goal is to install mercury removal units upstream the gas processing facilities.

Furthermore, mercury has also been found to be a serious ***poison to metal catalysts*** used in the reactions occurred during the hydrocarbon processing. To avoid the problems outlined above, all operators of LNG and many operators of conventional gas processing plants seek the total removal of mercury from natural gas and NGL plants.

As previously mentioned, an effective approach to achieve this goal is to install **Mercury Removal Units (MRU)** in gas processing units. These units are most often located at gas processing facilities that produce feedstock materials for down stream chemical manufacturing plants. The removal system can eliminate mercury from plant products and thus substantially reduces the impact of mercury on downstream plants. Gas processing plants vary considerably in design depending on the composition of the feed and the market requirements. Plants are optimized to make particular products such as LNG, LPG, NGL, ethane, propane, butane, and/or pentane (C_{5}-) depending on the feed to the plant and consumer market.

Mercury removal units are required for all LNG plants because of the sensitivity of cryogenic heat exchangers to mercury deposition; where the required low temperatures to liquefy gas usually condense mercury as well.

Furthermore, mercury can also cause ***severe health problems***, where exposure to mercury causes permanent damage to brain, nervous system, and kidneys.

Detection Methods and Limitations

The primary analytical methods for mercury in natural gas include colorimetric analysis, cold vapor atomic absorption sepectrophotometry, and amalgamation collection.

Colorimetric analysis can be conducted in one of two common procedures. Gas containing mercury can be passed through a potassium iodide or iodine solution, resulting in quantitative formation of mercuric iodide. A more common colorimetric detection technique is done with a Draeger tube. Gas is pumped through the tube which causes a chemical color changes as a copper/mercury complex is formed. The greater the volumes required for a complete color change reaction, the lower the mercury concentration. Draeger techniques are generally used only to detect high concentration of mercury (50 to 2000 $\mu g/Nm^3$).

In ***cold vapor atomic absorption sepectrophotometry technique***, mercury is reduced to an elemental state in an aqueous solution. The mercury is then volatilized, and absorption of ultraviolet radiation by mercury vapor is measured. Because of its low detection levels, this technique has become somewhat of a standard for mercury detection measurement.

Activated carbon and metals such as gold and silver (in the form of wire, wool, granules, and films) are used to collect mercury ***by amalgamation***. These collectors have excellent retention efficiencies, but various memory effects possibly resulting from diffusion of mercury atoms into inner layer of the collection medium. Consequently, the metal type collectors used now are extremely thin.

Gold film mercury detection is based on the concept that resistance increases linearly across the gold film as mercury is adsorbed. Measurement of this resistance change is a measurement of mercury mass.

Comparison between gold film and atomic absorption detection methods indicate that there is a linear correlation of the two methods with saturated ambient air samples. It was found that there are no significant differences between the results obtained by the analyzers except that the gold film system offers lower detection levels, and the determination of small quantities of mercury in natural gas by gold film detectors provides the most reliable results [10]

Conventional Mercury Removal Processes

Adsorption on Activated Carbon

The most widely used process for the removal of mercury is by its reaction with elemental sulfur to form mercuric sulfide.

$$Hg + S = HgS$$

The sulfur is first deposited on a support, typically active carbon, and the resulting captive mass is used in a standard fixed bed reactor. The reaction is rapid and high levels of mercury can be absorbed onto the bed. A number of drawbacks on this removal method were detected. Out of these drawbacks are the following;

- There is no commercial use for the spent material
- The only environmental acceptable way for disposal is by combustion followed by condensation of the evolved mercury
- Landfill is one of the most relied method of disposal

Adsorption on Sulfur Impregnated Carbon

The most commonly used method for removing mercury from natural gas streams is "chemi-sorption" on sulfur-impregnated carbon. This method is based on the principles of adsorption and chemical reaction of mercury present in natural gas using impregnated elemental sulfur in a micro-porous adsorbent. The adsorbent is primarily designed to extract elemental mercury, i.e. mercury

in its vapor state. The bed of sulfur impregnated carbon has to be located downstream of molecular sieve dryers and it can only be dried with cold gas.
The impregnation process increases the activated carbon capacity significantly, in order to produce a special type of sulfur impregnated activated carbon with a great adsorption capacity for mercury vapor. As a result, a stable and insoluble compound which is mercury sulfide is formed (10). The disadvantages of using this method were recorded as follows;

- The possibility for capillary condensation due to very small pore size
- Sulfur can be lost by sublimation and by dissolution in hydrocarbon liquids
- Operators' health problems due to the sulfur sublimation and condensation [9]

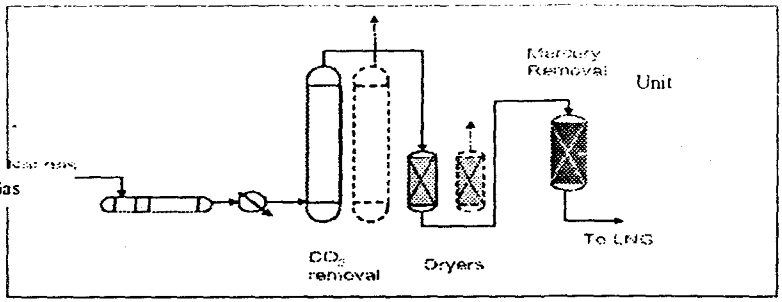
Amalgamation with A valuable Metal

It is also possible to remove mercury by amalgamation with a valuable metal such as silver. This provides effective removal and potential for regeneration, but it cannot work in the presence of high levels of H_2S.

Location of Mercury Removal System

Early designs of LNG plants used beds of sulfur impregnated carbon to remove mercury from the raw gas. Because of the capillary condensation problem, these beds are generally located at the final stage of purification after molecular sieves dryers, but it is not the ideal location. The different locations for the mercury removal system are:

- downstream from the dehydrator
- upstream the acid gas removal unit



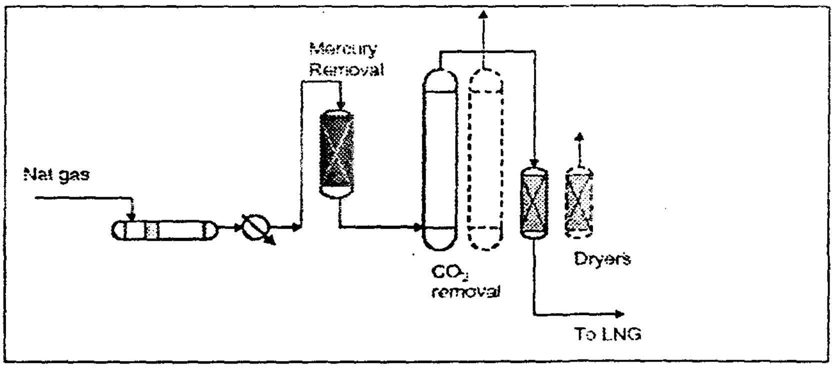


Figure (19): Location of Mercury Removal before Acid Gas Removal Unit

Factors Affecting Mercury Removal

Several factors affecting the removal efficiency of mercury from natural gas. These factors are summarized as follows:

- composition of gas
- presence of higher hydrocarbons
- operating temperature/pressure
- activated carbon characteristics
- concentration of mercury vapor

- presence of water and other impurities
- gas flow rate
- contact time[11]

Mercury Allowable Limits

Considerable amount of mercury is removed from the produced natural gas by surface adsorption or by condensation. The allowable concentrations of mercury should be reduced to certain values less than 0.01 μg/Nm^3 before the gas enters cryogenic process equipments.

Egypt Experience with Mercury in Natural Gas

As mention before, Egypt is currently implementing a challenging Liquefied Natural Gas Projects in Damietta and Idku. Although natural gas analysis for most of the gas fields and processing facilities is being performed on daily basis it is limited to gas constituents (C_1- C_6) in addition to nitrogen and CO_2 with no analysis for mercury existence.

Accordingly, it was necessary to benefit from the leading LNG experiences that were worldwide reported and take positive actions towards analyzing the gas taking the object of identifying level of mercury existence into account. Furthermore, it becomes a must to have a complete image regarding mercury in natural gas fields around the country especially that will be fed to LNG plants.

To achieve the above objects and others in the same concern, the Egyptian Natural Gas Holding Company is being sponsored to implement a research project entitled "Mercury Removal from Natural gas" in cooperation with the Academy for Scientific Research and Technology.

The project will extended over two years starting from October 2003. As per the project document, it is scheduled to achieve the targeted goals during four work phases as shown below.

Phase 1: Literature Surveys.

Phase 2: Preparation of necessary equipments for mercury trapping.

Phase 3: Measuring mercury levels in natural gas using Absorption Spectroscopic Techniques

Phase 4: Results/Recommendation and Final Report

The first phase is successfully finished ending up releasing the first project interim report. The report treated the following subjects;

- History of natural gas mercury problem.
- Natural gas general characteristics and processing.
- Natural gas profile in Egypt.
- Mercury overview, characteristics, occurrence, applications, Environmental cycle.
- Problems caused by mercury in natural gas.
- Mercury removal and trapping techniques.
- Atomic Fluorescence Spectrometer method used for analyzing natural gas mercury.
- Mercury detection in the Egyptian gas fields

The second phase is almost finished and the related interim report will cover the achieved activities within this phase. Out of the covered activities are the following.

- Identifying technical specifications for the proposed portable mercury analyzer.
- Configuring the Terms of Reference (TOR) for measuring mercury in natural gas fields.
- Surveying the pre-qualifications of the submitted companies for mercury determination.
- An overview on the suggested gas fields (geographical location, applied gas processing, possibility of gas sampling, etc).

The startup of the third phase will immediately begin with the specialized company that will take the responsibility of determining mercury in the suggested gas fields in co-operation with the project team.

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