A Review of Hydrate Formation in Oil and Gas Transition Pipes

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Abstract. This review provides a description of the various applications and issues of gas hydrate production in gas and oil pipelines, in general, gas hydrate is a clathrate of physical compounds, in which gas molecules are trapped in crystalline cells, formed by hydrogen bonds of water molecules. In the presence of water under various conditions, gas hydrates can be formed from all gases at high and lower pressures. The oil and gas industry has been taking stringent measures for many years to prevent the formation of hydrates which is plug pipelines by use. However, Natural gas hydrate is a non-conventional energy resource available to humans in colder regions, such as ice or sea bottoms. Other positive applications for natural hydrate include sequestration of carbon dioxide (CO2), storage, separation and transport of natural gas, the use of hydrate dissociation energy can be used in cooling and cool storage processes. The aims of this paper are to prevent gas hydrate’s formation that occur naturally in the oil natural gas transition pipelines, as well as the positive and negative that is remains from this natural phenomenon.

Keywords: Gas Hydrates, Synthetic Inhibitor, Modeling, Gas Transporting Pipe Lines

1. Introduction
Natural hydrates (e.g. water and gas) are crystalline solids. The gas molecules (guests) are trapped in the cavities of water which are hydrogen bonded water molecules (host) as shown in Figure 1 [1, 2] Composition (SI), Composition (SH) and Composition (SH) isometric. Lattice is. Three gas hydrate structures have been found. [1]. The cages of Structure I are organized in a body-Center , Wide enough for methane, ethane and alike molecular gasses, for example Carbon Dioxide and Sulfide Hydrogen. Packing is like a diamond in structure II, which results in some cages being big enough, not only to include Methane & Ethane gas, But gas molecules are as big as propane, Composition H takes a small. Molecule and also the larger molecules, typical of the gas or oil portion. Farther than these three, structure I is the most common one. Figure2 shows the three common structures of gas hydrate[3]

Gas hydrates are clathrates that develop under certain conditions (low temperature, high pressure) and may be present at increased temperature than the water freezing point. High pressure (typically > 30 bar) and low (typically < 20 ° C) conditions support hydrate formation. Precise conditions in terms of
pressure and temperature depend on the composition of the fluids. These conditions could exist offshore in shallow depths below the ocean floor.[4]

![Image](image_url)

**Figure 1.** Host molecules (water) and guests (gas) [5]

In 1934, Schmidt et. al [6] discovered that the clogging of natural gas pipeline was not caused by formation of ice, nevertheless by formation of Natural gas clathrate hydrates. This finding was a determining factor in increasing oil and gas companies’ interest. There has been a lot of work done to find prevention methods for the thermodynamic treatment of natural gas hydration network [7-10].

There are four method could use for avoiding hydration formation as show below:

1. Control of Pressure (The lower pressure is less hydration, but it is difficult in gas transportation lines because the gas pressure to move it is increased)
2. Control on Temperature (The system is heated by electrical heating in such a way as to prevent it from reaching the hydrate point of formation)
3. Remove the water (Water in the lines of the pipe should be removed. Despite this, there's always a little water in the gas)
4. Inject the chemical inhibitors (Such inhibitors block the development of hydrates and are used before other processes).

There will be two major groups of chemical inhibitors; thermodynamic and synthetic inhibitors; thermodynamic inhibitors affect the thermodynamic equilibrium of the aquatic phase. [5].

Also many industrial operations developed many techniques for avoiding gas hydrate formation such as natural gas can be desiccated; at a given pressure, The Hydrate formation equilibrium could be heated to the overhead temperature, Gas pressure can be decreased under the hydrate forming equilibrium Pressure at a certain temperature and the partial phase cycle of the gas-water system can be altered by injection into the hydrated region this called thermodynamic inhibitors of methanol, glycols or electrolytes. [11]

The physical properties of gas hydrates Nonflowing crystalline solids are more highly packed than traditional liquid hydrocarbons and gas molecules they produce, these properties give rise to numerous applications in the broad areas of energy and climate effects, such as separation technologies, due to high gas concentration [3].

Gas Hydrates can be used effectively, too for sequestration of Carbon Dioxide (CO₂) or for storage and transport of Natural gas. Furthermore, the hydrates of Natural gas occur Naturally in the bottomless sea, and high levels of permafrost So it turned out to be Potential energy source that has less influence on the environment. Lastly, Clathrate hydrates provide high latent dissociation heats which may be used for cooling applications, for instance cold storage or air conditioning [12]. On the opposite, gas hydrate was responsible for the plugging of natural gas pipelines [13]. In addition, gas hydrate formation during drilling in deep water can have several adverse consequences for well control and safe operations. [14]
2. Hydrate Basics

Three principal conditions are required to form gas hydrates. They are including:

1. The presence of a hydrate former – This is the gas or light hydrocarbon molecule that sits within the water cage and stabilizes the hydrate structure. Typical hydrate formers are methane, ethane, propane, carbon dioxide, nitrogen and hydrogen sulphide. However, some larger hydrocarbons can also form hydrates in the presence of these light gases.

2. Water is needed to form the hydrate cage; typically hydrates contain 85% water. The amount of water may affect the hydrate formation conditions. It is certainly thermodynamically possible for hydrates to form even when no free water is present – whether the hydrates actually form in these conditions or, if they do, form in sufficient quantities to cause a problem.

3. A suitable combination of temperature and pressure – Hydrate formation is more likely at low temperatures and high pressures. The actual temperatures and pressures will depend on the gas, condensate or oil composition but hydrates can certainly form above 0°C, the freezing point of water. Typical seabed temperatures are 4°C, well within the hydrate formation conditions for many gases, especially at high pressures. On-shore conditions can often reach these temperatures in winter months in many gas-producing regions. At 4°C many natural gases will form hydrates at less than 10 bars, while at 100 bar many common gases will form hydrates at ambient temperatures.

Zahng et al. [15] the conditions above are necessary for hydrate formation but there are many factors may favour hydrate production including:

- Nucleation positions: - This is a site which favors the formation of a new phase, in this case enhances the possibility of the first hydrate crystal forming from the fluid phase. In oil and gas production, such sites may vary from imperfections or disruptions in the smooth pipeline wall to the presence of other pipeline solids, such as sand particles

- Free water present: - Although not strictly necessary for hydrate formation, it is known that in many cases hydrates form at the gas-water interface. In gas-condensate-water systems, hydrates tend to form at the abundant gas-water interfaces, whereas in low gas-oil-ratio (GOR) gas-oil-water systems they tend to form at the oil-water interfaces. [16]

- Agitation: - Mixing process equipment encourages the production of hydrate by creating more interfaces between water and gas. When the stream passes through an impact, turbulence is the same and there is an increase in the probability of a sudden drop in temperature. [15]

2.1. Structure of Gas Hydrate:
After water molecules come into connection at low temperature and high-pressure gas molecules, a gas Hydrate ion formed of different geometrical structures that are contrary to hexagonal ice. The water molecules act as host molecules, creation of cage networks that can be molecules for guests, due to the presence of gas molecules, these crystalline cages similar to structures are a smaller amount dense than crystalline water structures. The gas hydrate formed by water molecules ’ hydrogen bonds is held together also stabilized by the forces of Van der Waals, holding together the gas and the liquid. Vander Waals forces are responsible for the hydrate's stable nature and make More stable hydrate than normal water ice. There are several gas hydrate structures, which are distinguished by their cage shape. Natural gas mainly composed of Methane gas and it is completely combustion provides water, Carbon Dioxide (CO$_2$) and energy, as shown in the Equation (1)

$$
\text{CH}_4 + \text{O}_2 \Rightarrow \text{CO}_2 + \text{H}_2\text{O} + \text{Energy} \uparrow
$$

(1)

The energy generated by this process can be used for various purposes. As more energy is released and CO2 is generated, natural gas is further environmentally convenient than other fossil fuels.

Figure 3 shows the structure of the gas hydrate. Methane gas is the middle (green) host, whereas the host (pink) is the water molecule.

As shown in Figure 4, Gas hydrate can be stored or transported at its saturation temperature or pressure under equilibrium conditions.

Generally, hydrates are stable at temperature and pressure of saturation. Some factors affect the capacity pressure and hydrate temperature, such as the cost and weight of material for the storage tank hydrate and the sediments ’ hydrate deposition environment. Hydrates are generally stable at medium temperatures and pressures relative with LNG and CNG specifications. [16, 17]

![Figure 3. Cage-like gas hydrate structure. [17]](image)
2.2. Gas Hydrate Structures Classification

The gases create from either biogenic or thermogenic sources in hydrates that occur naturally, biogenic gases are primarily products of methane microbial production. Hemogenic Gasses usually come from deep oil wells, and while methane is predominantly present in these gases, the hydrocarbon portion of (C2 - C5) is very important. In the case of thermogenic gas hydrate, the alteration in composition of the vapor liquid during this formation cycle is especially stimulating due to the valuable insight into the reaction mechanism. When methane gas is trapped, the different hydrate classes are formed. These are classified by cage structures, which are distinguished by the setup of water molecules in the crystals. Host molecules (H2O) are rearranged while the gas (CH4) is trapped, and a cage is formed to trap the gas. Not every molecule of water attracts a molecule of gas; Some of them remain empty even if a cage appears but, methane gas is released as soon as the gas hydrate falls. Table (1) shows the properties of the cages created by the three gas hydrate crystal structures. [18]
2.3. The structure and physical properties of natural gas hydrates

At certain time operating temperatures and under pressure conditions, the crystallizations that occur round the host molecules result in the raw multi-phase stream of natural gas. (NGH) [19, 20] and [21]. Methane, ethane, propane, I n-butane, nitrogen, carbon dioxide and hydrogen sulphide are the most commonly found guest molecules in natural gas mixture. Yet methane-based NGH is the most common among these. [6, 22-24], NGH consists of a guest molecule of around 85-mol percent; they have physical qualities therefore nearby to ice. The structure is crystal. The density of NGH depends on the former molecules and the conditions of formation [25]. Natural Gas Hydrates (NGH) is a part of a larger family of compounds named “Clathrates”, which is inorganic container compounds [26]. Although a large number of container and hydrate formers are found in crystal structures, this paper focuses on (NGH) formers and (NGH) structures.

Generally, Hydrates are classified according to the arrangement in the crystal structure of the water molecules. All common natural gas hydrates consist of three crystal structures as shown in Figure 5: cubic shape (SI), cubic shape (sII), hexagonal shape (sH). Shape (I) consist of guest molecules, For example: methane, ethane, Carbon dioxide, and hydrogen sulfide. Nitrogen and small molecules including hydrogen form shape (II) as single guests. single guest molecules such as propane or iso-butane will form structure II. Even larger molecules (typically 7Å < d<9Å) such as iso-pentane or neohexene may form structure H when accompanied by smaller molecules] methane, hydrogen sulfide or nitrogen. [27, 28]. The NGH crystal structures composed of water molecules are bound in a solid lattice with hydrogen. The interaction between the water molecules and the guests is very poor but the over-all interaction with the host structure is very strong. [29, 30]. More than 130 known cathartic compounds with water molecules are cited in literature and more attention is given to SI and SII hydrate since this is the NGH structure with a great deal of weight [31-34]. The (SH) structure of NGH is well defined [35-41]. Furthermore, the phases of high-pressure hydrate are studied [42-45]. Crystal properties of natural gas hydrate have been broadly studied[27, 32]
3. Gas Hydrate: Problems in Oil and Gas Industry

3.1. Gas Hydrate Occurrence during Drilling in Offshore Regions

Amodu [46] conclude that the solid Hydrate can plug well lines and cause well control problems, hydrate can form in riser and in the annulus between the casing and drill string, which may stop the circulation of drilling fluid and prevent the movement of drill string and seriously effect drilling operations as shown in Figure 6.

Ning et al [47] studied when the gas Hydrate Occurrence during Drilling in Offshore Regions, where note Increased problems with increasing water depth will be faced in offshore oil and gas exploration. Gas hydrates are one of these obstacles.

Liu Xiaolan and others [48] studied when shallow natural-gas sediments are reached during bottomless water exploration, this gas may enter the penetrating fluid, resulting in low temperature and high-pressure formation of the gas hydrate process. Also, If the drilling fluid used was not inhibited by hydrate, the gas hydrate could easily develop when the flow of the loam stopped and the gas entered the drilling fluid, this would lead to an accidental gas jump that would block the pipe during the drilling process.

Consequently, since the 1990s, Strict measures were taken to evade the formation of hydrates in bottomless-water penetrating by gas. The main protective measure is the addition of influenced chemical compounds that cause drilling fluids to prevent the formation of gas hydrates such as thermodynamic inhibitors and low-dose hydrate inhibitors (LDHIs). In the bottomless-water oil and hydrate drilling, thermodynamic inhibitors are normally added to the drilling fluid, while kinetic hydrate inhibitors (KHI) are still being investigated.
3.2. Issues of flow assurance

Oil and gas transport in the pipeline are a public method of carrying oil and gas from the well to the production site. If in the hydrate zone in the phase diagram the temperature and pressure of the pipe line falls, gas hydration particles start to form. Eventually these particles could plug the pipeline as shown in Figure 7. [49]

Esam Jassim [50] investigated experimentally that Hydrates form and clog the transmission pipelines, making uneconomic operation, stopping production, occasionally for as long as months, in large, extended pipelines. The distribution of hydrate appears to slowly shape a plug that splits the tube into two pressure sections: A high-pressure portion between the well or gas source and the plug; and a second low-pressure section between the plug and the gas recovery section. Due to increased pressure, a tube blast may occur at the high-pressure portion. If the pressure difference between the upstream and downstream sections decreases, the plug will break the pipe. The safety of staff and production equipment can be affected by both problems. [50]

Imen Chatti [12] investigate that So as to struggle hydrate plugs and ensure regular flow, four main processes are investigated: chemical additives, hydraulic, thermal and mechanical processes.

The chemical process can be classified into thermodynamic and low-dose hydrate inhibitors. Thermodynamic inhibitors reduce the movement of water by moving the hydrate phase down to lower temperatures and increasing pressure to avoid the effective formation of hydrated gas. Nevertheless, it is not uncommon in certain field cases to require high concentrations over 60 % Glycol and Methanol. The need for large volumes of Glycol and Methanol leads to high costs and increases logistical and environmental concerns. Low-dose hydrate inhibitors, on the other hand, are comparatively new classes of chemical addition which can be active and efficient at only a few mass percent for traditional applications. Low-dose hydrate inhibitors are classified into two groups of kinetic and anti-agglomerate hydrate inhibitors. Anti-agglomerates do not prevent hydrates from forming, but stop gas hydrates from agglomerating into non-moveable lumps. Kinetic inhibitors are chemical substances dependent on polymers. Kinetic inhibitors are not believed to prevent but delay the formation of hydrate. A standard concentration of approximately one percent of the mass is adequate to track the phase of Hydrate Formation till the gas or Oil is transported to the end point or processing facility where the hydrate formation is outside the thermodynamic circumstances [51]. The method of hydraulic removal is based on the process of depressurization separating the hydrate plug. Given the porous structure of the gas pipeline plugs, this method seems promising. Nevertheless, it is not suitable for liquid hydrocarbons because it is vaporized by depressurization. The thermal method consists of
the heat flow to the pipe wall to lift the device's temperature above the point of hydrate formation. This method is possible for external pipe lines but not suitable for subsea equipment [12]. In addition, mechanical pigging can be used to clean the pipeline for the deposit. The method involves moving a large disc or a spherical or cylindrical structure made of flexible material, e.g. rubber, and making the external diameter almost equal to the internal diameter of the tube to be cleaned, but it is costly to shut down production to perform this process.[52, 53]

Figure 7. Hydrate formation in natural gas transmission pipeline [49]

4. Clathrate Hydrates: Potential Applications

4.1. Natural Gas Hydrates as a Possible Source of Energy

Large amounts of methane gas hydrate occur in the form of solid in sediment and sedimentary rock within 2000 m of the earth’s crust in permafrost and deep-water regions Keith A. [54]. The quantities of gas hydrates in the crust of the Earth could be considered a new source of unconventional energy resource. It has been pointed out that under the same pressure and at the same temperature, Methane hydrate can contain a volume of 164 times as high as one of the methane gasses.[12, 55]

Potential reserves of gas in hydrate that distributed in offshore and on land are more than 1.5 x 1016m3 [56]

Gas hydrate plays an important role to satisfy worldwide energy needs, therefore the exploitation, development and pro-duction of gas from Gas hydrate is dedicate to be a distant prospect for the 21 centuries [57, 58]

Gas recovery is generally produced by either thermal, depressurization, and/or chemical inhibition. In order to release gas from a gas hydrate deposit by thermal stimulation one has to warm the formation through the injection of heated fluid or potentially direct heating of the formation. Depressurization is more preferable and inexpensive than the thermal stimulation method, because it does not require large energy expenditure and can be used to drive dissociation of a significant volume of gas hydrate relatively rapidly Carolyn [59]. However, it needs high porosity hydrate deposits and, at the transport period, the removed gas and water can re-crystal into Gas hydrates inside the transport pipelines and then cause the pipe to plug[12]. Chemical inhibition like certain organic (e.g., methanol and glycol) or ionic salts compounds, is used to change the gas hydrate stability condition [60]. Seawater salt or other chemical inhibitors may be required through certain stages of gas production from methane hydrate bonds, but would not be the primary means of extracting gas hydrate that has not been used for a long time or on a large scale.
4.2. **Carbon dioxide capture and sequestration:**
The impact of the industrial emission of carbon monoxide (CO2) and its effects on the environment leading to global warming has gained importance in the recent years [61]. The major source of Carbon Dioxide (CO2) releases in the atmosphere are thermal power generation, cement manufacture, steel and iron making, oil and natural gas refining, and petrochemical industries [62]. The carbon dioxide capture and sequestration (CCS) has become an important area of research in industrialized countries to overcome worldwide concerns over the threat of global warming [63]. Moreover, the basis of CO2 capture is to look at CO2 not only as pollution greenhouse gas, but also as an important raw material [62]. At this time available techniques for carbon dioxide (CO2) capture and separation are chemical solvents, adsorption, chemisorption, absorption, and chemical bonding through mineralization. Certain factors such as the quantity of chemicals used in this process, the energy legislations, and the prices related with these processes make these processes less attractive for large-scale carbon capture [63]. Therefore, Current, less energy-intensive architecture of systems is of major interest to research. The techniques of gas hydration crystallization have certain advantages, with water, which provides abundant (cheap) and green chemicals for carbon dioxide hydrating. In addition, The reduction of energy requirements for the formation of hydrate can be accomplished, including hydrate forming process, other organic chemicals in lower concentrations. [64]. After separation, the captured carbon dioxide must be definitely: sequestered. The main steps of CCS processes: capture, transportation, storage is shown in Figure 8. Many methods under study for CO2 sequestration, such as storage in depleted oil reserves, salt formations, terrestrial ecosystems, and geological formations or direct injection into the deep ocean [65]. Methods of carbon dioxide sequestration into the sea have been investigated for many years and a wide range of ideas has been considered. In bulk, gaseous carbon dioxide and sea water are compressed and transported as a solution in pipelines or in pressurized and chilled vessels and then pumped to the same pressure. The solution is then pumped downward to the depth in which the pressure and temperature is suitable for carbon dioxide hydrates formation. These hydrates sink towards the deep-sea bottom where, due to their density, they steady in the long term. [66, 67]

![Figure 8. The main steps of CCS processes: capture, transportation, storage](image)

4.3. **Advantages of Gas Hydrate**
Azeez G. [18] studied and show the advantage of Methane hydrate, its commonly called gas hydrate, there is a huge quantity of energy that could be used for various purposes. Natural gas mainly contains
colorless, odorless and combustible methane gasses for the generation of Carbon dioxide (CO2), water and important energy levels. A lot of permafrost and bottomless-water marine Gas hydrate deposits. Gas hydrate deposits can be traveled for the production of methane gas as a maintainable energy resource. Gas hydrate can be used as an alternative to flaring gas from a viable natural resource, as a way to store and transport natural gas.

4.4. Natural Gas Storage and Transportation:
The gas hydrate is used for natural gas transports and processing as hydrates have the ability to provide high gas concentrations. [64]. Several studies show that the gas hydrate structures have a potential as storage media for various gases. Natural Gas hydrate (NGH) is considered to be a significant technology among the many potential technologies for carrying gas from production to use. Pipelines include: liquefied natural gas (LNG), compressed gas (CNG), liquid gas (GTL), commodity gas (GTC) pipeline. The economic advantage of storing and transporting natural gas in the form of gas hydrates is primarily due to reduced investment in infrastructure and equipment. [68, 69]
The key to NGH storing and transport is to overcome longer induction periods and improve hydrate formation. Many researches have been done to increase the rate of formation of hydrate, including adding surfactants to the solution, stirring, and bubbling. Nonetheless, the economic aspect remains the separation factor for optimum system output determination. [68, 70]

4.5. Applications of cooling storage
The increasing demand for electrical power for residential air conditioning and ozone depletion by chloro-fluorocarbon (CFCs) emphasizes the use of alternative cool storage systems that shift this demand to an off-peak period, eliminating the need to use conventional refrigerants (CFCs) and the need for HFC (hydrofluorocarbons) [71]. Carbon monoxide (Co2) Hydrate is an alternative way of using clathrate hydrate slurries as a two-phase (solid-liquid) refrigerant in the cooling process. These two-phase coolants are more energy efficient than single-phase refrigerants due to high latent fusion heats. As in the case of ice slurries, direct gas injection into a watery solution will produce heat dissociation of CO2 hydrates. The heat dissociation of these slurries was found to be appropriate for cooling. [71, 72].

5. Natural Gas Hydrate Hazards & Safety:
The safety factor of the NGH is taken into consideration in gas processing plants and sites experiencing NGH issues; storage and transport of pipelines. Since the hydrate crystal structure, It has a high latent energy. It requires melting before explosive gas leakage is released during an accident, inherently preventing LNG and CNG from leaking into the hydrate system. In terms of storage, NGH slowly burns and does not fire when ignited. Therefore, once the hydrate carriers’ walls are broken down, natural gas hydrates are not readily discharged from the carriers, as is the case for LNG. Nevertheless, with the presence of a hydrate plug in the transport pipelines in mind, there are greater safety issues, especially when the hydrate plug is removed. When the hydrate plug is disconnected in the pipeline, the pressure drops when the pipeline is stripped from the pipe wall. This pressure difference may result in high mass solid hydrate plug inside the pipeline. This will further reduce the downstream gas, resulting in pipeline leakage, breakage and damage. [73-76] Hydrates contain a lot of gas volumes at standard temperature and pressure per volume of hydrate. Separation of hydrates by heating causes a rapid increase of gas pressure in the system A pipe may be broken by attempts to blow a plug. Knowledge of the position and length of a hydrate blockage is very important to determine the best approach to hydrate blockage remediation. There may be several plugs in the pipe that threaten the pipeline, from safety and technical, during plug dissociation. As inspected by The Canadian Association of Petroleum Producers there are two points to consider in that regard [48, 74].
- Always assume several plugs of the hydrate in the flow loop. Between two plugs there are high pressure value.
- Trying to push hydrate plugs will split Pipes and containers in the flow chain.
Depressurization may potentially start a plug like a high-speed bullet that causes the setting to trigger broken pipes, damaged equipment, and uncontrolled release of hydrocarbons.

6. Conclusion
The purpose of this literature review is to distinguish between different areas of interest related to the advantages of gas hydrates or hydrate problems in oil and gas industry where, gas hydrate is a regard of important process which can happen naturally, gas hydrate has advantage and disadvantages: first can be used in many applications like source of energy, gas separation from the flue gas mixture, and cool storage application, Second At the same time, it is regarded as a problem in the oil and gas industry due to the formation of gas hydrates during gas and oil drilling in offshore, and in gas and oil transporting pipe lines which causes many problems in finances, although there are four methods to avoid hydration effects such as (controlling pressure, controlling temperature, removing water, and injecting chemical inhibitors), so conclude that hydrate formation in the gas transport pipe line was based on hydrate growth velocity with inhibitor decreases and this decrease is different based on the type and density of the inhibitor used. This paper gave various areas of interest related to gas hydrate advantages or hydrates problems in oil and gas operations.

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