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Gas hydrate formation in H_2–natural gas mixtures: Insights for energy

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ABSTRACT

Hydrogen is a clean and versatile energy carrier with significant potential for supporting a carbon-neutral future. Blending hydrogen with natural gas may improve energy performance, but it also introduces challenges - particularly the risk of altered gas hydrate formation. Hydrate plugs can severely impact pipeline operations, making it vital to understand how hydrogen affects hydrate behavior. This study presents original laboratory results using a custom-designed apparatus that simulates hydrogen-natural gas mixtures under realistic pressure and temperature conditions. The system ensures precise control of hydrate formation, and the findings provide valuable insights to support the safe and efficient integration of hydrogen into existing gas infrastructure.

KEYWORDS

green hydrogen, decarbonization, hydrogen hydrate, renewable energy, gas hydrate

1. INTRODUCTION

The transition to renewable energy sources is a central goal in addressing global climate change and meeting international environmental commitments. Among the many benefits of renewable energy is its ability to reduce greenhouse gas emissions associated with the burning of fossil fuels. Despite the significant increase in the share of renewables in the global energy mix, fossil fuels - namely coal, oil, and natural gas - are still projected to account for over 76% of total energy production by 2040 [1].

Hydrogen, especially when produced from renewable sources (so-called "green hydrogen"), has emerged as a clean, versatile energy carrier with applications across multiple sectors, including transportation, industry, and energy storage [2]. However, hydrogen's role in balancing energy supply and demand is especially promising. Because energy consumption fluctuates daily and seasonally, excess energy generated at low-demand periods can be stored as hydrogen and reused later. One method under consideration is the storage of hydrogen in hydrate form.

Hydrogen can be stored in several ways: compressed gas cylinders, absorption into materials with high surface area, or chemical bonding with host materials [3, 4]. Among these, hydrogen hydrates - formed by trapping hydrogen molecules within water-based crystalline structures - have gained attention due to their safety, environmental compatibility, and relatively low cost. As it is shown in Fig. 1, hydrate-based storage is part of a growing research area aiming to overcome the limitations of traditional hydrogen storage technologies [5].

This paper presents an experimental study on hydrogen hydrate formation under simulated pipeline conditions. A custom-built testing apparatus was developed to examine the effects of varying hydrogen concentrations in natural gas on hydrate behavior.

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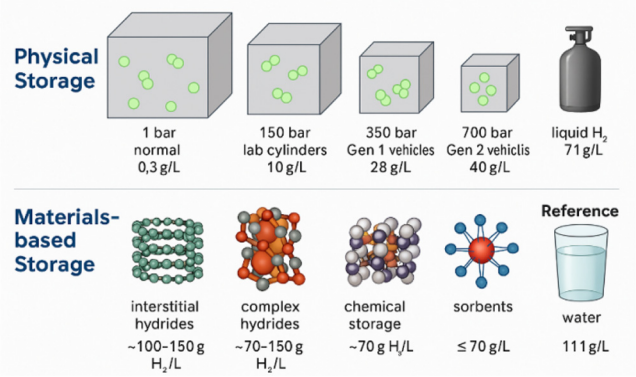

Fig. 1. Storing hydrogen in physical storage vs. storing it in materials (*Source*: figure recreated by the Authors based on data from [5])

The experimental results provide new insights into the mechanisms of hydrate formation in hydrogen-enriched systems and support the development of safe and efficient hydrogen storage and transport solutions for future energy infrastructures.

2. FUNDAMENTAL CONCEPTS AND METHODS

2.1. Forming hydrates

Gas hydrates are crystalline compounds in which small guest molecules become trapped within a lattice of hydrogen-bonded water molecules. These structures form under specific thermodynamic conditions - typically low temperature and high pressure - and can store large volumes of gas in a compact solid form. When temperature increases or pressure decreases, the hydrate decomposes, releasing the gas in its original form [6].

The phenomenon of gas hydrate formation has a long history, dating back to 1778, when Joseph Priestley [7] observed sulfur dioxide bubbling in cold water. Later, in 1810, Humphry Davy [8, pp. 27–41] described the formation of clathrate hydrates, though the subject remained relatively obscure for over a century. The phenomenon of gas hydrate formation has been known for centuries, with early observations dating back to the late 18th and early 19th centuries [7, 9]. However, scientific interest in the topic remained limited until the 1930s, when operational problems in the growing US oil and gas industry - for example, pipeline blockages occurring during cold winters - were traced to hydrate formation. It was during this period that the role of water vapor in forming solid hydrates under specific conditions was first recognized [7].

This discovery triggered systematic investigations into gas hydrates, which remain relevant challenges in the energy sector today. In the 1960s, naturally occurring gas hydrates were discovered in the permafrost of Yakutia at a depth of 1,200 m [10] - marking the beginning of the third major phase of hydrate research. Since then, over 7,000 scientific articles have been published on the subject, exploring their implications for both energy production and environmental safety [7, 9].

More recently, researchers have explored the use of hydrates in various energy-related applications, including hydrogen storage, carbon capture, and natural gas transport. Initially, hydrogen was considered unsuitable for hydrate formation due to its small molecular size and low polarizability [3]. However, in the 1990s, Dyadin et al. [11] demonstrated that at pressures above 100 MPa, hydrogen could indeed form hydrate-like structures in ice matrices. These findings were further supported by the work of Lokshin et al. [12] and Mao et al. [13] who studied hydrogen hydrates in high-pressure environments.

Although these high-pressure hydrates represent a scientific breakthrough, their practical use is constrained by the extreme conditions needed for their formation. As a result, researchers have turned their attention to semi-clathrate hydrates formed with compounds like Quaternary Ammonium Salts (QAS), which present a more feasible alternative. These structures remain stable at ambient temperature and pressure and have demonstrated the ability to host hydrogen molecules within their cavities [14]. As a result, they represent a promising direction for developing safer and more efficient hydrogen storage technologies.

2.2. Hydrate's ability to store hydrogen

One of the most attractive characteristics of gas hydrates is their ability to store large amounts of gas in a compact solid form. The volume of gas in hydrate form is significantly smaller than in its free gaseous state. For example, at 15 °C and $1.01 \cdot 10^5$ Pa, a gas mixture containing 80% CH_4, 10% C_2H_6, 5% C_3H_8, 4% n-butane + isobutane, and 1% inert gases occupies 4.42 m^3. In contrast, the same amount of gas stored as hydrate requires only 1/156 of that volume. This compactness is a major advantage, especially when space and safety are key considerations.

In addition to space-saving, hydrate storage offers important safety benefits. Hydrates are non-flammable, and their decomposition into gas can be carefully controlled, which significantly reduces the risk of explosion. This makes hydrate-based systems particularly appealing for storing and transporting flammable gases like hydrogen.

There are ongoing efforts to develop efficient methods for hydrogen storage in hydrate form. Although promising, the practical use of hydrogen hydrates is currently limited by the need for very high pressures and low temperatures to form and maintain the hydrate structures. For example, the highest hydrogen storage capacity reported for sH hydrates is approximately 4.2% by weight [15]. However, when promoters are added to facilitate hydrate formation, they often occupy hydrate cavities themselves, reducing the overall hydrogen capacity.

To enhance storage potential, researchers are exploring ways to improve hydrate stability and hydrogen density by optimizing pressure and temperature conditions. Increasing pressure and lowering temperature have been shown to improve hydrate formation kinetics and storage efficiency, particularly for sH-type hydrates. Figure 2 illustrates the filling behavior of different hydrate structures as a function of pressure at 200 and 300 K [16].

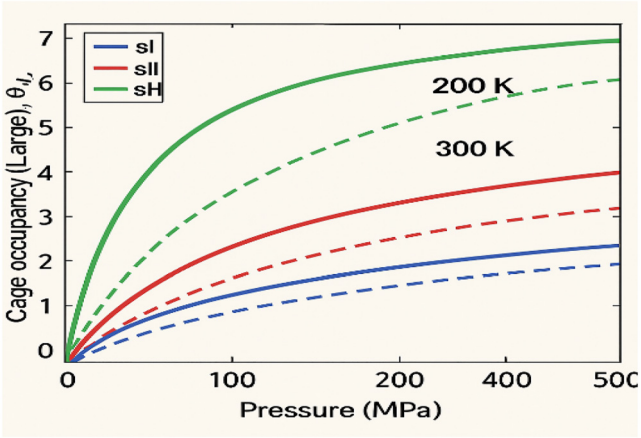

Fig. 2. Filling of the cavities corresponding to the three hydrate forms as a function of pressure at 200 and 300 K (*Source*: figure recreated by the Authors based on data from [16])

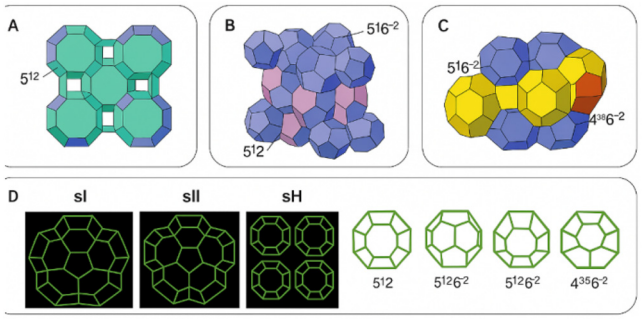

Fig. 3. Structures of hydrogen hydrate (*Source*: figure recreated by the Authors based on data from [17])

2.3. Structure of a hydrogen hydrate

Clathrate hydrates are inclusion compounds where guest molecules are trapped within a crystalline lattice formed by hydrogen-bonded water molecules. These structures are stabilized by van der Waals interactions between the water framework and the enclosed gas molecules. Hydrate structures are typically categorized into three types: structure I (sI), structure II (sII), and structure H (sH), each with unique cage geometries and compositions.

- sI hydrates contain 46 water molecules per unit cell, forming two types of cavities: large cages with two hexagonal and twelve pentagonal faces, and smaller cages with twelve pentagonal faces;
- sII hydrates consist of 136 water molecules and have small cages formed by twelve pentagonal faces, and larger ones with twelve pentagonal and four hexagonal faces;
- sH hydrates, the rarest of the three, are composed of 34 water molecules. Their structure includes small cages with twelve pentagonal faces, medium cages with a mix of pentagonal, hexagonal, and tetrahedral faces, and large cages with twelve pentagonal and eight hexagonal faces [17].

The stability of these structures depends heavily on the size and shape of the guest molecules. As noted by Berecz and Balla-Achs [9], very small and weakly polarizable molecules - including hydrogen, helium, and neon - do not easily form hydrates. However, research conducted by Dyadin and colleagues in the 1990s [15] demonstrated that at sufficiently high pressures, hydrogen can indeed be incorporated into hydrate structures, paving the way for high-pressure hydrogen storage.

The formation and stability of hydrates can be further described through cohesive and interaction energies, which quantify the strength of hydrogen bonding within the structure. Figure 3 presents a comparative view of the three hydrate structures in both 2D and 3D formats, illustrating their geometric differences [17].

3. NATURAL GAS HYDRATES ANALYSIS

Studying the formation and dissociation of clathrate hydrates at the molecular level presents significant experimental challenges. To address this, researchers have increasingly turned to Molecular Dynamics (MD) simulations, which allow for detailed investigations of hydrate nucleation, growth, and structural behavior under various thermodynamic conditions.

MD simulations help uncover the mechanisms and properties of hydrate formation, from nucleation behavior and structural types to mechanical stability. Several algorithms have been developed to identify and analyze hydrate cage structures:

- Face-Saturated Incomplete Cage Analysis (FSICA): Recognizes both complete cages and partially formed ones, making it effective for identifying hydrate clusters and multi-guest systems [18];
- CHILL+ (CHILL Plus) is an improved algorithm that determines whether a water molecule belongs to a liquid or solid phase by evaluating its orientational order relative to its nearest neighbors. This method builds upon the original CHILL (Crystalline Hydrogen-bonded Ice-like Lattice Locator) algorithm [19];
- GRADE: (Graph-based Ring and Cage Detection Engine) is an algorithm that uses a Breadth-First Search (BFS) to map ring connectivity in hydrate cages. It identifies five-membered rings and merges them based on their shared edges [20];
- Iterative Cup Overlapping (ICO): Efficiently detects standard edge-saturated cages with minimal computational cost and high accuracy [21];
- Hydrate Topological Recognition (HTR): Utilizes unique topological pathways formed by intersecting rings to identify hydrate cages rapidly and reliably [22].

These algorithms support detailed structural analysis, which is critical for designing and evaluating hydrate-based energy systems. Figure 4 illustrates the operational principles of each algorithm, demonstrating their role in advancing hydrate research.

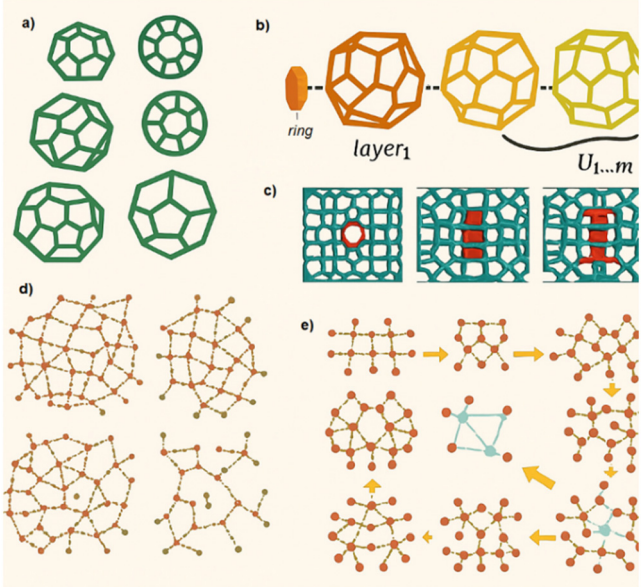

Fig. 4. Algorithms for hydrate cage structure identification use identification principles (*Source*: figure recreated by the Authors based on data from [23])

4. APPLICATIONS OF NATURAL GAS HYDRATES

Experimental studies and molecular dynamics simulations of gas hydrate formation offer valuable insights for various practical applications. One of the most actively investigated areas is the potential of gas hydrates as a future energy source. Methane hydrates, in particular, are recognized as a promising alternative to conventional fossil fuels due to their high energy density and widespread availability. It is estimated that methane hydrates contain more carbon than all other fossil fuels combined and are evenly distributed across the globe, both in permafrost regions and under the ocean floor [24].

The economic and environmental impact of utilizing natural gas hydrates may even surpass that of shale gas, as they represent a vast and relatively untapped energy reserve. According to [25], hydrate reservoirs offer significant energy potential with gas concentrations up to 164 times the original hydrate volume and dissociation that consumes less than 15% of the recovered energy. These features make gas hydrates a valuable resource in the transition to cleaner energy systems.

However, hydrogen-enriched gas mixtures may pose challenges related to internal corrosion in pipeline infrastructure. Studies focusing on inhibitors like NALCO® EC1005A provide valuable insights into preserving system integrity under these conditions [26].

Another critical application of hydrate research is the prevention of hydrate formation during the transport of oil and gas, particularly in subsea pipelines located at depths exceeding 1,800 m. At these extreme conditions - characterized by low temperatures and high pressures - hydrates are likely to form, which can result in pipeline blockages, operational interruptions, and environmental hazards.

Traditionally, thermodynamic prevention methods - including heating, insulation, dehydration, and the use of chemical inhibitors - have been employed to reduce the risk of hydrate formation. However, these techniques are often expensive and demand specialized expertise and infrastructure. Advances in hydrate modeling and experimental research may enable the development of more cost-effective and environmentally sustainable solutions for managing hydrate-related risks in the energy sector.

5. MEASUREMENT

The experimental setup used in this study is a dynamic hydrate testing apparatus developed by researchers at the University of Miskolc, in collaboration with experts from a leading Hungarian oil and gas company. This custom-built equipment was adapted specifically for testing hydrogen–natural gas mixtures and simulates pipeline conditions found in real gas fields.

The system enables small-scale testing that corresponds to typical gas well outputs and pipeline dimensions. The following design parameters were applied when building the miniature test loop:

- Inner pipe diameter: 2 mm;
- Pipe material: carbon steel;
- Pipe length: 6 or 12 m;
- Gas pressure: 60 bar;
- Gas flow rate: 4 nL min^{-1}.

Using this configuration, processes occurring between the wellhead and the collection station can be realistically modeled. In operation, a hydrogen-methane mixture is passed through the test pipe, and a controlled amount of brine is added to initiate potential hydrate formation. The pipe section is placed in a liquid thermostat, allowing precise temperature control. Due to flow resistance, pressure differences develop across the pipe, which are measured to detect hydrate formation.

The test equipment includes a pre-cooling unit, which consists of a liquid thermostat with adjustable temperature, an insulated container, internal cooling pipes, a temperature transmitter, and differential pressure sensors. The apparatus allows tests in the temperature range of −20 to +30 °C and simulates field-like pressure conditions. Gas flow can be adjusted between 1 and 10 nL min^{-1} to match different modeling scenarios.

The equipment's P&I diagram (see Fig. 5) includes the following key components:

Sz-01 is the pressure reducer, Sz-02 to Sz-08 are the on/off valves, PU-01 is the brine pump, TC-01 is the temperature control, MV-01 is the magnetic valve, TT-5 is the temperature transmitter, PT-1 to PT-2 are the pressure transmitters, PT-3 to PT-4 are the differential pressure transmitters, FT-6 is the mass flow transmitter, GP-01 is the

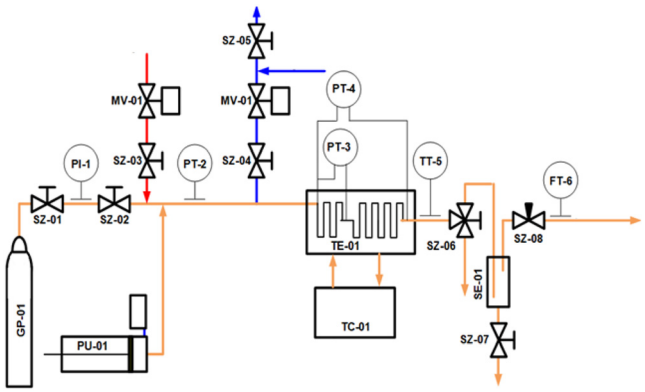

Fig. 5. Test equipment P&I drawing (*Source*: figure created by the Authors)

gas tank, SE-01 is the separator cell, and TE-01 is the thermostat.

In future development, integrating AI-based calibration methods described in [27], may enhance sensor reliability in field applications.

During the experiments, the test temperature was set at 0 °C, the gas flow rate at 4 nL min^{-1}, and the system pressure at 58 bar.

Hydrate formation was induced by gradually increasing the amount of brine introduced to the hydrogen–methane mixture. Hydrogen concentration levels were varied 0%, 5%, 10%, 15%, and 20%, and for each concentration, the brine flow rate was adjusted incrementally from 0.7 to 2 mL min^{-1}. Measurements recorded the timing of cooling, the start and end of the test, the number of data points, and whether hydrate formation occurred.

A clear trend was observed: higher hydrogen concentrations required more brine and longer formation times for hydrates to develop. In some cases, no hydrate formation occurred at all, even with increased brine dosage.

Table 1 presents a summary of the measurement results across various hydrogen concentrations. It includes the test date, hydrogen content, brine flow rate, timing of the cooling and measurement phases, the number of recorded data points, and whether hydrate formation was observed.

Table 1 provides a comprehensive overview of the experiments conducted with varying hydrogen concentrations and brine flow rates. The table records important test parameters, including the exact timing of the measurement phases and the number of data points collected. This allows for a nuanced understanding of hydrate formation under different gas compositions.

The data reveal several clear trends:

- At 0% hydrogen, hydrate formation occurred consistently across all tests. Even minimal brine injection rates 0.7–0.8 mL min^{-1} were sufficient to trigger the formation of solid hydrates in the test section. Hydrate plugs formed quickly, as seen from the relatively short measurement times and sharp pressure increases;
- At 5% hydrogen, hydrate formation became less predictable. In one case (2021.08.30), no hydrate formed at all, despite a typical flow rate. In other tests, increasing the brine flow rate to 0.8 or 1.0 mL min^{-1} restored hydrate formation. This indicates a threshold behavior, where slight changes in hydrogen content or brine quantity significantly affect the outcome;
- At 10% hydrogen, the inhibition effect becomes more pronounced. Two tests failed to produce hydrates, even at higher brine levels. Only when the brine rate reached 1.2 mL min^{-1} did hydrate formation resume. These findings suggest that hydrogen, when mixed into natural gas, acts as a moderate thermodynamic inhibitor;
- At 15% and 20% hydrogen, hydrate formation was largely suppressed. Despite brine rates as high as 2.0 mL min^{-1} and long measurement durations, no hydrate plugs formed in most tests. Only one test at 15% hydrogen with 1.5 mL min^{-1} brine produced detectable hydrate formation.

This implies that hydrogen content above 15% may effectively prevent hydrate nucleation and growth, possibly

Table 1. Examination results

Date	H2 content (%)	Brine (mL min^{-1})	Cooling	Start time (hh:mm)	Shutdown time (hh:mm)	No. of measurement points (pcs)	Hydrate
2022.04.11	0	0.70	9:00	10:28	11:17	615	yes
2022.04.13	0	0.75	9:35	10:38	10:56	230	yes
2022.04.12	0	0.80	9:25	10:06	10:24	233	yes
2021.08.30	5	0.60	12:10	12:48	14:13	1,067	no
2021.07.28	5	0.80	9:35	10:44	11:32	608	yes
2021.07.21	5	1.00	10:00	10:49	11:05	200	yes
2021.09.02	10	0.80	10:30	11:08	12:21	920	no
2021.09.08	10	1.00	11:30	12:47	14:30	1,304	no
2021.10.14	10	1.20	10:20	11:07	12:08	771	yes
2022.03.01	15	1.20	11:10	11:59	12:24	320	no
2022.03.03	15	1.32	9:20	11:02	12:38	1,192	no
2022.03.09	15	1.50	12:20	14:05	15:14	836	yes
2022.03.17	20	1.50	12:15	13:51	15:00	881	no
2022.04.06	20	1.80	10:25	11:20	13:25	1,249	no
2022.04.07	20	2.00	8:10	8:50	10:36	1,319	no

due to its molecular size and inability to stabilize the hydrate cages.

Figure 6 presents the result with 0% hydrogen. The graph shows two differential pressure sensors (PT3 and PT4) registering sharp increases during the test. This pressure buildup corresponds to hydrate plug formation in the test section. The steepness of the pressure curves suggests rapid nucleation and growth once the conditions were met.

Figure 7 presents the result with 5% hydrogen. Here, pressure buildup occurs more slowly, indicating that more time or brine was needed to reach hydrate-forming conditions. The final plug formed was smaller, and the curves rise less steeply than at 0%.

Figure 8 presents the result with 15% hydrogen. Although pressure readings show some fluctuation, they do not reach the levels seen in lower-hydrogen tests. This suggests partial or unstable hydrate formation, likely insufficient to create a full blockage. The hydrate structure may have begun forming but failed to reach the critical mass needed for propagation.

Figure 9 presents the result with 20% hydrogen. No significant pressure change is observed. Despite high brine input and extended test duration, no hydrate plug formed. This confirms the strong inhibitory effect of high hydrogen concentrations.

The results demonstrate a clear inverse relationship between hydrogen concentrations and hydrate formation

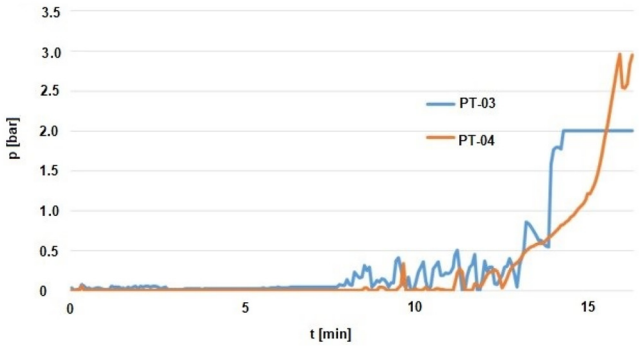

Fig. 6. Result with 0% hydrogen (*Source*: Figure created by the Authors)

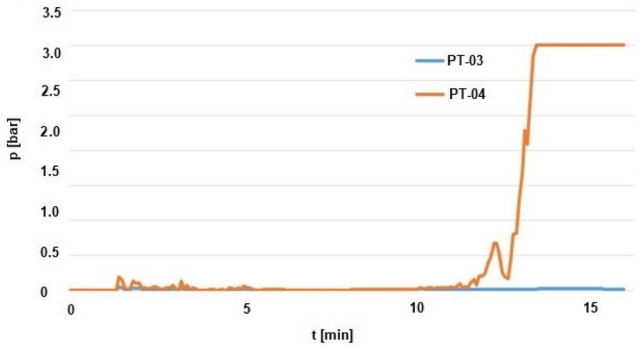

Fig. 7. Result with 5% hydrogen (*Source*: figure created by the Authors)

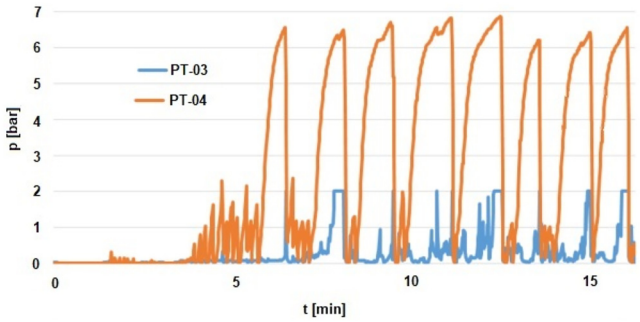

Fig. 8. Result with 15% hydrogen (*Source*: figure created by the Authors)

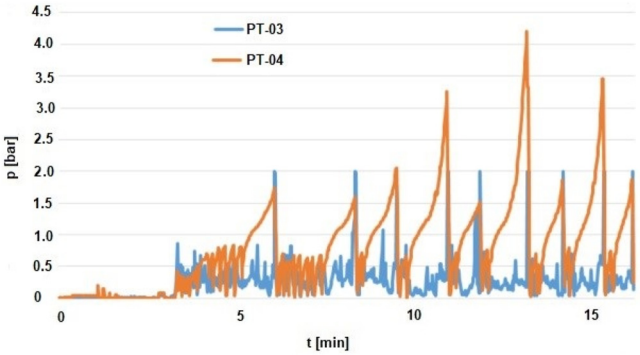

Fig. 9. Result with 20% hydrogen (*Source*: figure created by the Authors)

likelihood. While small amounts of hydrogen (up to 5–10%) only slightly delay formation, higher levels (15–20%) can effectively inhibit hydrate development altogether. This has two important implications:

Operational safety and cost savings: If hydrogen-enriched natural gas naturally inhibits hydrate formation, the use of expensive chemical inhibitors (e.g., methanol or monoethylene glycol) could be reduced or eliminated.

Hydrogen injection limits: Understanding these interactions is essential for determining safe hydrogen blending ratios in gas networks, especially under high-pressure, low-temperature conditions common in gas transport pipelines.

6. CONCLUSION

According to forecasts, by 2050, humanity's energy demand is expected to triple compared to the turn of the millennium, making the exploration of alternative energy sources both urgent and indispensable. One of the key steps in moving away from fossil fuels is the drastic reduction of greenhouse gas emissions, primarily carbon dioxide.

The results of this study demonstrate a clear inverse relationship between hydrogen concentration and the likelihood of hydrate formation. While low hydrogen content

(5–10%) only slightly delays hydrate development, higher concentrations (15–20%) can effectively suppress it altogether. This finding is particularly significant, as commonly used hydrate inhibitors - including methanol and monoethylene glycol - are both costly and environmentally harmful. Therefore, blending hydrogen into natural gas offers both economic and environmental benefits by potentially reducing or eliminating the need for these chemical additives.

Future research will focus not only on transportation aspects but also on storage conditions for hydrogen–natural gas mixtures. To this end, rock permeability tests will be conducted using prior experience, in order to gain a realistic understanding of how this new energy carrier interacts with reservoir rock. This knowledge is essential for the safe integration of hydrogen into gas infrastructure, especially under the high-pressure and low-temperature conditions typical of gas transport systems.

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