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Origin and significance of methane hydrates and their generation under laboratory conditions

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Abstract

Purpose. The purpose of this paper is to present a conceptual design for laboratory-scale equipment for the controlled generation and investigation of small quantities of methane hydrate under variable pressure, temperature, and lithological conditions, enabling reproducible experimental studies that simulate natural methane hydrate formation environments.

Methods. The study includes a review of recent global research and pilot-scale activities related to methane hydrate exploration, occurrence, and production. Key geological and thermodynamic conditions governing methane hydrate formation are summarized. Based on this background, an engineering concept for a laboratory apparatus is developed, including a high-pressure chamber, a gas compression system, and a temperature control unit, designed to operate within the methane hydrate stability zone.

Findings. The proposed apparatus is compact, modular, and suitable for installation in standard university laboratory conditions. It allows controlled variation of pressure and temperature parameters required for methane hydrate formation while maintaining operational safety and repeatability. The design relies on commercially available components and can be assembled with relatively modest financial and technical resources.

Originality. The presented solution is a practical, cost-effective alternative to complex, expensive laboratory systems commonly used for methane hydrate research. Its originality lies in integrating accessible industrial components into a simplified experimental setup capable of reproducing key conditions for methane hydrate formation in controlled laboratory settings.

Practical implications. The proposed laboratory system provides researchers, educators, and students with a flexible experimental platform for studying methane hydrates in water or porous media. It supports interdisciplinary research in geosciences, energy engineering, and environmental studies, and can be effectively used for academic training and experimental research.

Keywords: *methane hydrates; gas hydrate stability zone; unconventional gas reservoirs; laboratory equipment*

1. Introduction

Methane hydrates are well-known unconventional energy sources with vast potential reserves worldwide. One cubic meter of gas hydrate releases, at the surface, 164 cubic meters of methane. The present reserves are still unknown, because they have not been produced anywhere, but the probability ranges are extensive, from 7000 to $20000 \cdot 10^{12}$ cubic meters [1]. There are many sources on this topic, but the most comprehensive, including real datasets, is currently available on the website of the U.S. Department of Energy (DOE) Fossil Energy and Carbon Management (FECM) Gas Hydrates R&D Program [1].

The future commerciality of such sources depends on numerous variables, but the most important will be the price of natural gas. However, now is the right time to develop research and development procedures for such prospects, mainly because they could be in three very different environments: the ocean floor (offshore), mostly weakly compacted sediments below it, and permafrost (onshore). This makes potentially recoverable resources a challenging target, especially for drilling and recovery.

To date, several test fields, i.e., reservoirs, have been developed to the production phase, including extensive exploration and experimental testing. A famous example is the 2017 Gulf of Mexico Drilling and Coring Expedition, led by the University of Texas (Austin), where much of the data and results were linked to publicity [2]. The projected "methane hydrate reservoir" was drilled into a sandy and clayey silt bed, from millimeter to meter scales in Green Canyon Block 955 in the deep-water Gulf of Mexico (belonging to the levee of a turbidite channel). The methane saturation with hydrate (99.99% of gas) ranges from 79-93% in sandy silts, and is absent in clayey silts, when the origin of methane is mainly biogenic [3]. The authors noted that some sediment properties, with salinity close to that of ocean water, suggested that hydrates formed either long ago or recently, but slowly. Conversely, hydrate formation increases salinity. Sandy silt with its corresponding permeability could be an economically cost-effective reservoir for methane hydrates [3].

There is also the example of the Iġnik Sikumi gas hydrate experimental field, explored by ConocoPhillips in partnership with the U.S. Department of Energy, Japan Oil, Gas and

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Metals National Corporation, and the U.S. Geological Survey within the Prudhoe Bay Unit on the Alaska North Slope during 2011 and 2012. The goal was to determine the feasibility of gas injection to hydrate saturated sediments and to make a cost-effective exchange of injected CO_2 or a mixed CO_2-N_2 (more favourable) for produced CH_4 [4]. The methane flow was first observed at pressure above the methane stability zone (MSZ), and later it persisted for 30 days as the pressure dropped below MSZ. The authors [4] concluded that the destabilization of MSZ is self-limiting; i.e., uncontrolled destabilization is not expected, and sand production can be mitigated, as in other reservoir types. In addition, tests must be carefully performed to prevent wellbore blockage during operations stoppage, and reservoir heat exchange was sufficient to prevent wellbore freezing.

Testing production was also realized in Japan, after the first attempt in 2013 to produce in the Daini-Atsumi Knoll, the eastern Nankai Trough area near Honshu Island, a new trial was conducted in 2017 at a nearby location using two production and two monitoring wells [5]. The authors noted that the 1st well recovery lasted 12 days, with a stable drawdown of around 7.5 MPa and 41000 m^3 of methane. Later, the 2nd well operated for 2 days, and 222500 m^3 of methane was recovered without inflow of the sand particles from the reservoir. Due to higher water production, drawdown was limited to 5 MPa. Although Yamamoto et al. [5] emphasized that the energy produced from methane hydrates was significantly greater than the energy spent on production operations, they also noted weaknesses, such as a continuous reduction in the volumes of produced gas over time and significant differences in volumes produced between the 2 wells.

Zhu et al. [6] mentioned that five hydrate accumulations have been discovered in China, namely in the Shenhu, Dongsha, Qiongdongnan Basins, offshore of the South China Sea and in the Muli area of Qilian Mountain, adding that seven others have been assumed elsewhere. The authors estimated methane resources in hydrates to be $64.6 \cdot 10^{12}$ m^3 in the South China Sea, $\sim 28.5 \cdot 10^{12}$ m^3 in the East China Sea, and $\sim 38 \cdot 10^{12}$ m^3 in terrestrial permafrost, which is about twice the methane resource estimates in conventional reservoirs.

Selected examples showed that the exploration and exploitation of potential methane hydrate reservoirs can be time-consuming and financially intensive, requiring numerous pieces of drilling, logging, and laboratory equipment. Consequently, such ventures are available only to large commercial or academic participants with resources and expertise in hydrocarbon exploration and production. However, there is a niche where laboratory experiments and equipment can be used relatively cheaply to observe methane hydrate behavior under different temperatures, pressures, and lithological conditions. For example, Livio [7] noted that hydrate formation kinetics in porous media is a significant factor in revealing their formation history. The formation kinetics in silica sand environment, modelled with small cores and investigated in custom-designed equipment, was presented. The author also examined different CH_4 injection flow rates and eventually simulated geological conditions under three clay/sand lithologies. The idea for creating relatively inexpensive, yet precise and adaptable laboratory equipment for studying methane hydrates in water or rocky environments is presented as the main topic of this paper.

2. Occurrence, origin and migration of methane hydrates

Methane hydrates are icy solids that consist of methane and water. They belong to a group called clathrates, in which one molecule forms a cage that encloses another. If the cage-forming molecule is water, it is called a hydrate. If the molecule trapped in the water cage is a gas, then it is a gas hydrate, in this case methane hydrate [8]. They are stable only at pressures above 35 bar and/or at low temperatures within the methane stability zone (MSZ) or gas hydrate stability zone (GHSZ). The ocean floor is the ideal environment for their formation, with temperatures within 1-4°C. Below a water depth of about 350 m, the pressure is sufficient to stabilize the hydrates. However, below the ocean floor, in sediment, temperature begins to rise again due to the influence of the geothermal gradient, e.g., at depths greater than 1 km, temperature can exceed 30°C.

On most continental sea shelves, which are mostly shallow [9], significant volumes of methane hydrates have not been detected or predicted for two reasons: (a) the pressure at the bottom is not so high to create the hydrates and (b) the generation of organic matter and subsequent deepening at higher temperatures and compaction are not sufficient for the thermogenic generation of hydrocarbons, even methane. This second reason, the starving environment for significant quantities of organic matter deposition, and relatively fast sinking and compaction, is also crucial since large ocean basin areas deeper than 2000 to 3000 m (bathyal and abyssal) do not host any significant methane hydrate volumes. Their waters are nutrient-poor [8], and there is an influx of clastics. Even such a poor organic influx must pass through a large column of water to reach the ocean bottom, where many other living organisms could consume its organic remnants.

As De Haas et al. [10] noted, the role of shelves as C_{org} sinks is sometimes overestimated. Most continental shelves will not favor or even preserve a significant accumulation of organic matter due to several factors, like higher depositional energy (a lot of gravel and sand detritus that are easily eroded and destroy organic remnants), oxygen level and UV radiation (in the epipelagic, euphotic or sunlight zone). On shelves, significant quantities of organic matter and C_{org} accumulation can be preserved and buried only locally. Thus, continental slopes, canyons and deep-sea fans can be considered as the main depositional and preservation areas for C_{org} [10].

So, methane gas is primarily formed thermogenically from organic matter buried in fast-sinking sediments through phases of dia-, cata- and metagenesis onto slopes. Minor volumes can be generated biogenically by living microorganisms in relatively shallow, mainly marine sediments of terrestrial origin (e.g., in prodelta environments or similar settings). The methane gas generated deep within the sediment (in source rocks or biogenic) migrates upward and (part of it) is caught in methane hydrates, due to the decreased temperature in sediment pores or water near the sea floor. It is essentially a process of free thermogenic or biogenic gas migrating into and through the GHSZ. Several mechanisms were proposed to describe this process, including the inhibition of hydrate formation at higher salinity and temperature, and the identification of [11] chimney-like structures as favorable sites for gas migration from reservoirs below the GHSZ. The authors Liu et al. [11] described such structures as places where hydrate generation also raises the temperature within them, making hydrate accumulation favorable

close to the ocean floor, where temperature decreases again at bottom water values, producing a salinity peak at such locations. As migration and creation slow down, the temperature and salinity drop. Such a cycle can be repeated whenever methane from sediments is generated in larger quantities. In addition, hydrate migration can have a significant horizontal component, as Davies et al. [12] modelled for the Mauritian margin, where migration of more than 40 km below the GHSZ has been proposed, with venting through 23 pockmarks on the shelf. Interestingly, such events were triggered by past Quaternary interglacial episodes of warmer climate.

Consequently, methane hydrates form mainly on the continental slopes at water depths between 350 and (rarely) 5000 m, where organic matter type and quantity, as well as temperature and pressure, are favorable for methane conversion into methane hydrates [8]. The partial exception could be the Mediterranean Sea [13] and the Black Sea [14], [15], given their sizes and sedimentation rates in their deepest parts. In the southeastern Mediterranean Sea, the Levant Basin, GHSZ is determined through numerical modelling and seismic interpretation. The water column is more than 1 km thick, with an average temperature of 13.8ºC and salinity of 38.8‰. In such conditions, the top of the GHSZ was predicted at approximately 1250 m [13]. Taking into account the geothermal gradient in the sediments 20-28.5ºC/km, the same authors predicted the spreading of the GHSZ across the NW half of the basin, including about 150 m of sediments below the sea floor. Another example of a continental sea where GHSZ has been proven is the Black Sea. The sediments at depths of 500-600 m, saturated with methane, are potential reservoirs with reserves of about $100 \cdot 10^{12}$ m^3 [14], where hydrates are included into rock (15-60%) intercalations.

Vast volumes of methane hydrate are buried in sediment or ocean floors, comprising somewhere between 1000 and 5000 gigatonnes of carbon bound in hydrate deposits, which could be equivalent to 100 to 500 times the amount of carbon emitted into the atmosphere from fossil fuels [8]. The influence of methane hydrates on the climate can be predicted using various mathematical models. However, these models yield wide ranges due to many uncertainties in Earth's ocean and atmospheric systems and their mutual interactions. In most models, the methane hydrates located directly at the boundaries of stability zones (MSZ) will be affected primarily. Generally, such zones include hydrates formed in the water column above ocean floors (Fig. 1), at depths of approximately 450-700 m [12], and shallower in the permafrost above the Arctic Circle (Fig. 2).

Interestingly, most of methane released at the ocean floor does not reach the atmosphere (some estimates suggest that about 3.5% can vent into the atmosphere [12]), but is mainly converted into carbon dioxide, consuming part of the ocean's oxygen. Such CO_2 is then partially dissolved into the ocean, increasing acidification, and the rest is released into the atmosphere. The degradation of methane is very intensive during its upward migration through sediments below the ocean floor and involves two biological processes: anaerobic and aerobic oxidation, both catalyzed by bacteria [8]. Anaerobic conversion, under ideal conditions, can proceed through several chemical reactions, yielding calcium carbonate ($CaCO_3$). Later, such carbonate can be incorporated into sediments and rocks. However, the aerobic process is much riskier due to the potential impact of released methane as a greenhouse gas.

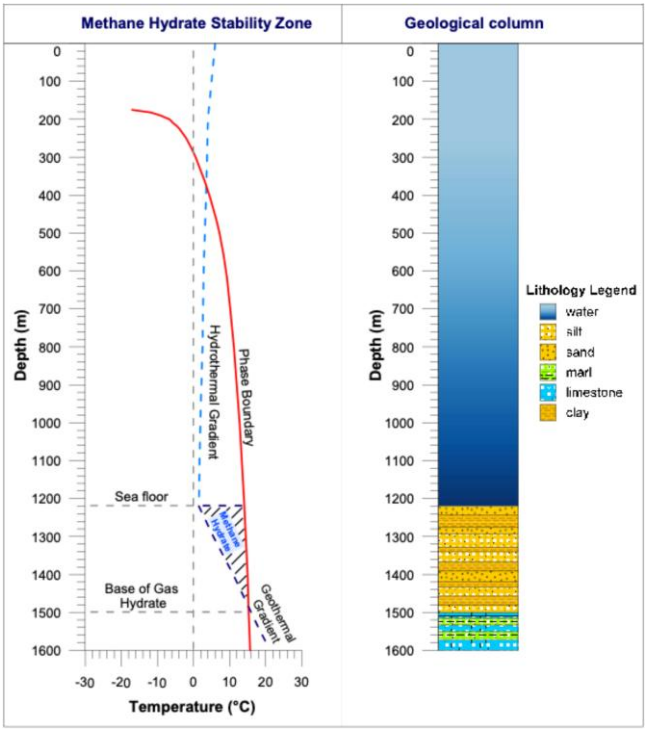

Figure 1. Example of the GHSZ in the water column above the ocean floor and sediments immediately below (prepared according to [16])

Much of the methane is degraded by bacteria using oxygen into carbon dioxide. It leads to ocean acidification and reduced oxygen levels. Some estimates show that about 90% of methane released from hydrates is consumed by those two processes before reaching the surface, and efficiency is increased if methane migration through a water column is slower [8]. It is worth noting that if methane release is abundant and rapid, it will not dissolve in water, since the bacteria will not have time to act, and methane will be released to the surface in the form of bubbles. This type of migration is also more probable in a shallow water column, where dissolution is weaker or absent, i.e., on continental shelves or top slopes. In addition, the role of methane hydrate in sediments as a cement may be important for assessing the stability of primarily such weakly consolidated rocks if methane has been released.

The threat of methane-related climate risk can be reduced through controlled production. Until now, there is no commercial field where such hydrates are produced economically, but volumes of methane in such hydrates exceed the proven reserves of natural gas (about 95+ % of methane) in conventional and other types of unconventional reservoirs. Some sources [8] predicted that methane hydrates could be economically feasible (as oil-equivalent reserves) with oil prices topping 50-60 USD per barrel. Some authors also emphasized the artificial production of methane hydrates during (subsurface) methane production, such as in coal mines [17], as a means to rationally utilize methane from coal mines, thereby reducing gas emissions. The authors experimentally determined favorable variables for hydrate generation at 1°C, pressure 10 MPa and duration of 2.5 hours, pointing out that the larger the methane concentration mixed with air, the greater the pressure required for hydrate generation.

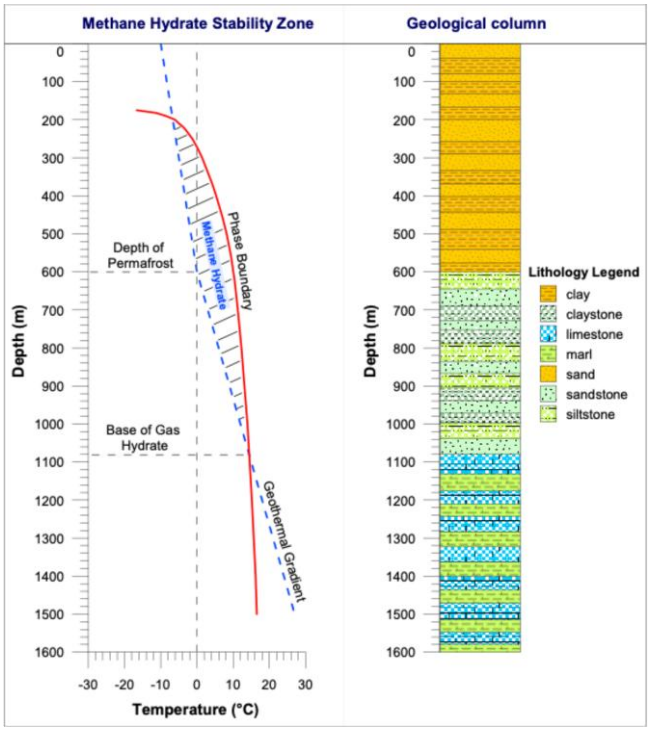

Figure 2. Example of the GHSZ in the deeper Arctic permafrost and sediments immediately below (prepared according to [16])

3. Conceptual model of laboratory equipment for methane hydrate production

Methane hydrates, beyond their academic interest, are among the primary future energy sources. In addition to natural conditions where they are accessible by drilling, it is helpful to study them in a laboratory under controlled conditions. To date, most research on methane hydrates and their behavior under potential reservoir conditions has been conducted using geological-numeric models and by testing at several sites where their trial production was attempted. The U.S. Department of Energy website hosts the largest and most well-known public database, including several case studies [1]. However, most existing studies address only the theoretical concept of the formation of such hydrates under different environmental conditions, but do not offer guidance on how to produce and study this chemical compound under relatively standard laboratory conditions and in small quantities as needed. Only a few such successful attempts have been described, primarily in large professional laboratories, such as those operated by hydrocarbon exploration companies or on drilling rigs at potential reservoir sites [18]. However, both cases are sporadic and require significant investments. After a comprehensive search on the Internet and the fact that hydrates can also be formed in the presence of gases such as ethane, propane, butane, or carbon dioxide, only one video describes how to create not methane but propane hydrate, due to the inability to establish the conditions necessary for the formation of methane hydrates, primarily pressure, using small and inexpensive laboratory equipment [19]. Additionally, there are only a few manufacturers of ready-made laboratory equipment for producing hydrates under laboratory conditions, while the prices are available only upon request.

Today, methods for high-speed synthesis of gas hydrates are rapidly developing, but existing laboratory equipment for producing methane hydrates under controlled conditions is prohibitively expensive. Consequently, it remains inaccessible to many research teams with limited financial resources, which have sufficient academic knowledge and want to expand it through experimental work. The high cost of laboratory equipment also poses a barrier for many scientific research institutions that are interested in methane hydrates. This limited access, due to high costs, highlights the need for more affordable approaches that enable a broader range of research teams to perform experimental work with methane hydrates. Furthermore, producing methane hydrates under controlled conditions is challenging because the conditions required for their formation are difficult to achieve.

In some papers describing the synthesis of methane hydrates using expensive laboratory equipment, pressures of up to 160 bar were applied [20], [21]. However, the literature states that methane hydrates begin to form at 30 bar and the appropriate temperature [22], [23]. To achieve the mentioned conditions, individually or together, considering the critical values for methane hydrate formation, it is necessary to develop specific equipment for such purpose, which is the objective of the conceptual model presented in this paper.

In addition to ensuring the presence of the previously mentioned thermodynamic parameters necessary for methane hydrate formation, a technical challenge of this conceptual model is the reliable control of these parameters. Since the minimum pressure required to form a methane hydrate is already relatively high, the chamber must be capable of withstanding that pressure and higher pressures, at which methane hydrates will form when combined with an appropriate temperature. For safe operation with methane within the specified pressure range, certain valves must be integrated into the chamber. Furthermore, an additional technical challenge is the need to visually monitor the methane hydrate formation process to confirm its formation. For expensive laboratory equipment, this is provided by endoscopic chambers integrated into the chamber interior. In addition, for better hydrate formation kinetics under controlled conditions, in the case of laboratory equipment, a magnetic stirrer is integrated into the interior of the chamber to increase the contact surface between the two phases, water and gas (methane). Another challenge is to ensure the repeatability of the methane hydrate formation process. Most commercially available methane sources are not compressed to the pressure required for methane hydrate formation. Therefore, methane must be additionally pressurized before it is injected into the chamber. If methane is already stored at the pressure required for methane hydrate formation, as is the case with certain gas cylinders filled with methane, the main issue is the pressure drop within the cylinder after several hydrate formation cycles. Consequently, at some point, methane from these sources must be additionally compressed outside the cylinder before being injected into the chamber. Repeatability is essential, as producing the required quantities of hydrates enables subsequent experiments to be conducted to investigate and understand the phenomena that occur during deep drilling to discover or develop potential deposits, such as assessing the impact of fluid filtrate reducers on methane hydrate formation when drilling with water-based muds [24], [25].

To effectively solve the listed technical problems, the solution for the chamber, based on our conceptual model, is a high-pressure coupling with integrated side elbows for a pressure gauge, a check valve, and a bleed valve. At both

ends, the coupling is closed by the sight glasses, of which the upper one provides the visual confirmation of the methane hydrate formation process. In addition, to ensure the repeatability of the methane hydrate formation process, a solution is to apply a gas booster. Our conceptual model does not include a magnetic stirrer to improve the methane hydrate formation kinetics, since there is information in the literature that methane hydrates can be formed without mechanical mixing of the two phases [26]-[28], i.e. water and methane, with the emphasis on the fact that mixing only affects the hydrate formation kinetics. In the case of such a design and in order to accelerate the process of methane hydrate formation, kinetic promoters can be added to water [29] or a multipurpose metal material made of copper with a network of open and closed pores in the form of copper foam can be used [30]. To achieve the temperature required for methane hydrate formation, the internal tank of the floor-standing water dispenser can be converted into a cold bath, cooled by the dispenser's existing cooling system, which is controlled by a temperature regulator.

A comprehensive internet search we conducted also showed that, according to the design of our conceptual model, all chamber components can be purchased online from companies that sell standardized, certified products in technical fields and industrial equipment. The accompanying equipment covered by this conceptual model can also be purchased from the appropriate websites, and the prices of both the chamber components and the accompanying equipment are publicly available and affordable. For this reason, a professional with knowledge of mechanical engineering, including welding, can build the chamber himself. To ensure the safe handling of the chamber, the primary objective is to ensure that the threaded connections of all chamber components are compatible with one another to provide adequate sealing and prevent leaks at high pressure. Chamber components can also be manufactured in a certified workshop that provides manufacturing services for products in the technical field, in accordance with the customer's specifications and dimensions.

The chamber within this conceptual model of laboratory equipment is flexible in its design. It can also be made in a monolithic shape, with the only detachable element being the upper sight glass, while all the other components form a single unit. However, for easier replacement of components in the event of damage, for example, any threaded connection due to long-term use, a more suitable solution is a chamber made up of several separate components. Since our approach involves a chamber that can be manufactured in a certified workshop and because the accompanying equipment described in our conceptual model is publicly available, our invention constitutes a safe and affordable technical system for producing methane hydrates under laboratory conditions. Figure 3 shows the chamber components.

When manufacturing the chamber in a certified workshop, the dimensions of the chamber components, in millimeters, are provided in the cross-sections of Figure 4. These dimensions ensure the chamber fits inside the internal tank of a floor-standing water dispenser. Comparable dimensions of chamber components are also available on relevant websites.

All chamber components must have the same maximum working pressure, which is important for the chamber's integrity. Our selection for the chamber material would be stainless steel class 3000.

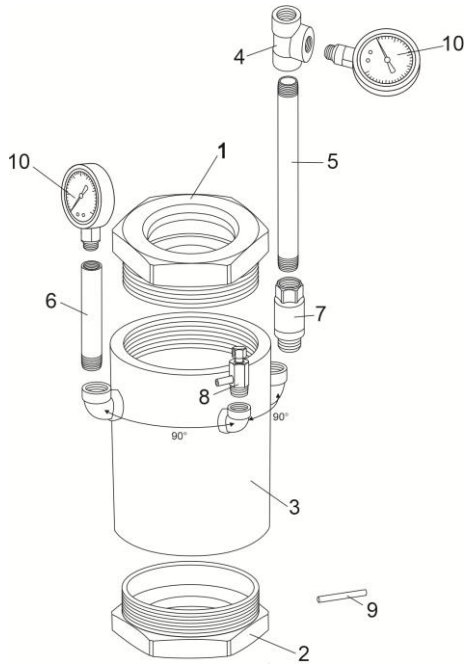

Figure 3. Chamber components: 1 – upper sight glass; 2 – lower sight glass; 3 – high-pressure coupling with integrated side elbows; 4 – T-adapter; 5 – longer pipe nipple; 6 – shorter pipe nipple; 7 – check valve; 8 – bleed valve; 9 – bar handle; 10 – lower mount pressure gauges

This class comes under a high-pressure class and is used for higher-pressure service conditions. It also indicates that the chamber can withstand pressures up to 3000 psi (206.84 bar). Although such pressure will never be achieved, thus, the safety factor is built in. This is also important because, as the temperature in the stainless steel class 3000 chamber increases, its pressure-holding capacity decreases slightly. Although methane hydrate synthesis is exothermic, the mentioned effect is expected to be negligible, as the cold bath should perform as required.

In addition, Figure 3 shows that threaded connections interconnect all components of the chamber. In general, pressure systems use different sealing methods depending on the type of system and environmental conditions. For example, many pressure systems in the oil and gas industry use National Pipe Thread (NPT) threads. However, there are other thread types, such as British Standard Pipe Taper (BSPT), British Standard Parallel Pipe (BSPP), National Pipe Taper Fuel (NPTF), and others. At the same time, regional differences and industry preferences also determine the type of threads used. If chamber components are manufactured in a certified workshop, the best type of threaded connection should be selected in consultation with the workshop's experts.

Certain thread types, for example, National Pipe Taper (NPT), typically require a thread sealant, such as Teflon tape, a gasket, or an Oring, to ensure a leakproof connection. If a sealant is not used with this type of thread, small gaps can allow leakage, especially with high-pressure gases. Therefore, for the mentioned type of thread, a high-quality methane-compatible thread tape is necessary. In addition, when applying Teflon tape, only two turns are recommended, as excessive application can lead to leaks. In contrast, the previously mentioned National Pipe Taper Fuel (NPTF) threads are designed to provide a more leak-free seal without the use of Teflon tape or other sealants.

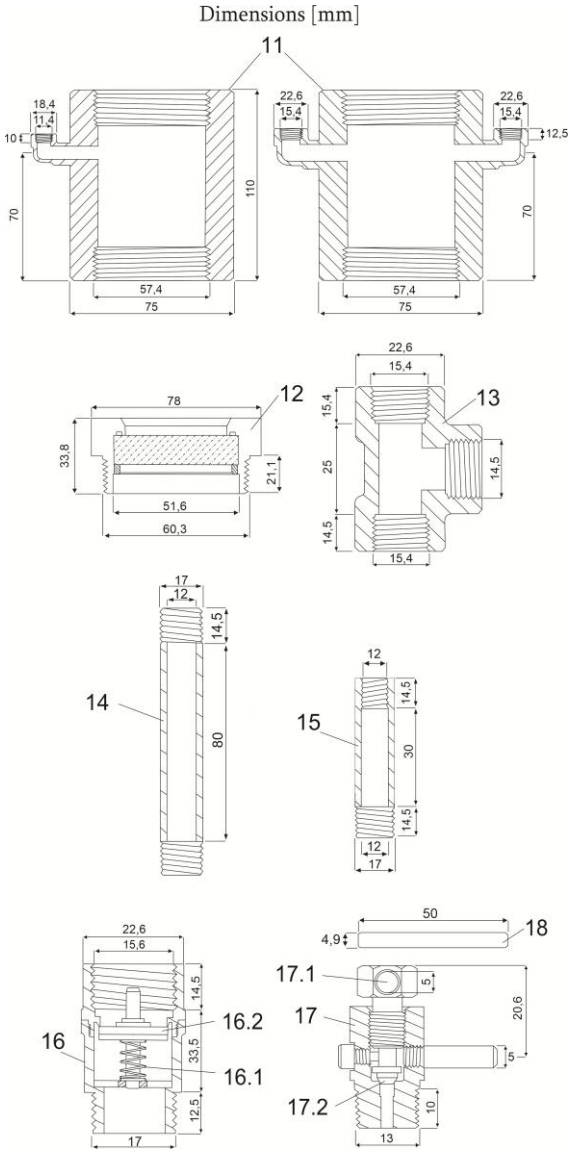

Figure 4. Cross-sections of the chamber components and their corresponding dimensions in millimeters: 11 – cross-sections of a high pressure coupling with integrated side elbows; 12 – cross section of a sight glass; 13 – cross-section of a T-adapter; 14 – cross-section of a longer pipe nipple; 15 – cross-section of a shorter pipe nipple; 16 – cross-section of a check valve; 16.1 – spring; 16.2 – plate; 17 – cross-section of a bleed valve; 17.1 – rounded opening; 17.2 – screw; 18 – cross-section of a bar handle

The National Pipe Taper Fuel (NPTF) threads are designed to crush together as they are tightened, creating a metal-to-metal seal. This design provides an enhanced seal without the need for additional sealants. The primary advantage of National Pipe Taper Fuel (NPTF) threads is their ability to prevent leaks at the thread roots and crests, even under high pressure. This self-sealing property makes National Pipe Taper Fuel (NPTF) threads ideal for high-pressure applications where a reliable seal is essential. When purchasing chamber components from relevant websites, the main issue is the adjustment of the male and female threaded connections on all components. Since male and female thread connections on all chamber components must use the same thread type, this approach requires careful consideration of the available thread types for male and female connections.

Since this chamber design does not include a magnetic stirrer, Figure 5 shows the process of filling the chamber with a mixture of water and a kinetic promoter.

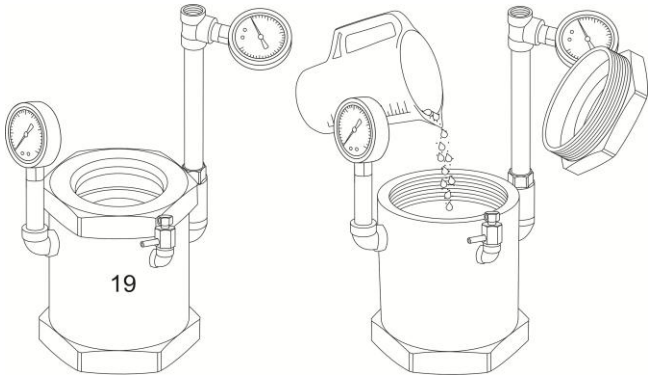

Figure 5. The procedure of filling the chamber with a mixture of water and a kinetic promoter: 19 – chamber

At the beginning of this chapter, it was noted that the solution to the technical problem of achieving the pressure required for methane hydrate formation and ensuring process repeatability is the use of a gas booster. Therefore, Figure 6 illustrates the methane injection process.

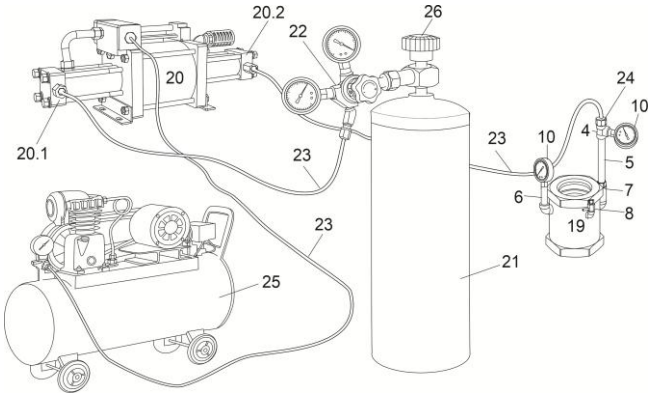

Figure 6. Methane injection under high pressure into the chamber using a gas booster and accompanying equipment: 4 – T-adapter; 5 – longer pipe nipple; 6 – shorter pipe nipple; 7 – check valve; 8 – bleed valve; 10 – lower mount pressure gauges; 19 – chamber; 20 – gas booster; 20.1 – inlet section; 20.2 – outlet section; 21 – low-pressure methane gas cylinder; 22 – pressure regulator; 23 – high pressure hoses; 24 – male-threaded quick connector; 25 – air compressor; 26 – cylinder valve

A low-pressure methane gas cylinder is connected to the gas booster inlet section through a pressure regulator and a high-pressure hose. On the other side, the gas booster outlet section is connected via a high-pressure hose with a male-threaded quick connector to the upper female-threaded port of the T-adapter (Fig. 6). An air compressor drives the gas booster and is connected to its air drive section via a high-pressure hose. The gas booster working principle is based on the pressure ratio between the gas outlet pressure and the set air drive pressure. This means that for a given air drive pressure, the gas booster delivers much higher methane pressure at its outlet. By opening the cylinder valve at the top of the low-pressure methane-filled gas cylinder, methane enters the inlet section of the gas booster (Fig. 6). When air from the compressor enters the air drive section of a gas booster, a system of pistons (larger and smaller) inside the booster

begins to move (Fig. 7). The movement of the larger piston, driven by air, is mechanically transferred to the smaller piston, which, through two stages of compression, compresses the methane to the pressure required for the formation of methane hydrate. Continuous operation is achieved via a pilot valve. At the end of the first stroke, the pilot valve switches the air supply to the opposite side of the larger piston, causing it to move in the opposite direction. During the return stroke, the smaller piston, driven by the larger piston's motion, compresses the methane again, thereby achieving the required pressure. The outlet pressure of the methane is directly related to the set air drive pressure of an air compressor. Methane from the outlet section of a gas booster is then injected into a chamber via a high-pressure hose (Fig. 6).

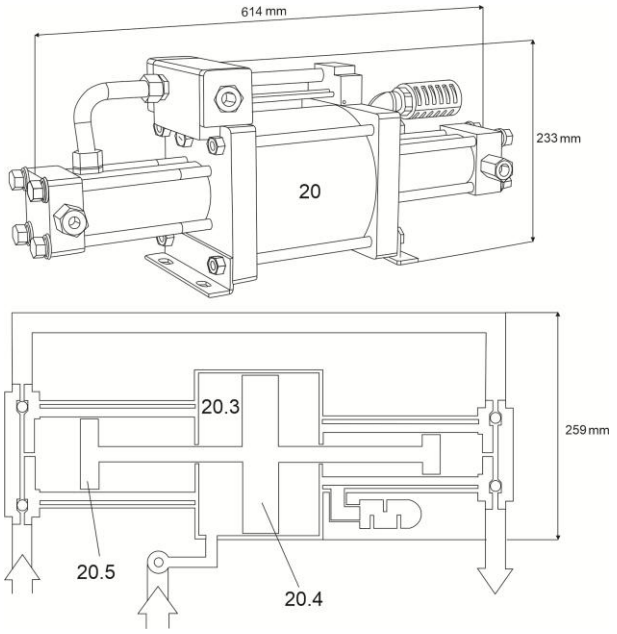

Figure 7. Two-stage, one air drive section gas booster working principle and its dimensions in millimeters: 20 – gas booster; 20.3 – air drive section; 20.4 – larger piston; 20.5 – smaller piston

During methane injection into the chamber, the check valve is open, allowing methane to enter the chamber (Fig. 6). A pressure gauge screwed into the female-threaded side port of the T-adapter allows monitoring of the methane pressure just before it passes through the check valve and enters the chamber. When the needle of the pressure gauge, connected to the shorter pipe nipple, shows a predefined pressure value, the cylinder valve at the top of the gas cylinder filled with low-pressure methane can be closed, and the air compressor can be turned off. At this injection stage, the pressure within the longer pipe nipple is lower than the chamber's internal pressure. As a result, the pressure in the larger pipe nipple is too low to overcome the spring force in the check valve, so the check valve closes, preventing methane from returning from the chamber. Throughout the entire methane injection process, the bleed valve remains closed. Since the chamber is designed as a closed system, and both the check and bleed valves are closed, methane is trapped under the required pressure and cannot escape from the chamber. If necessary, the chamber can be disconnected from the injection apparatus shown in Figure 6. After methane injection, the chamber must be immersed in a prepared cold bath inside the floor-standing water dispenser (Fig. 8).

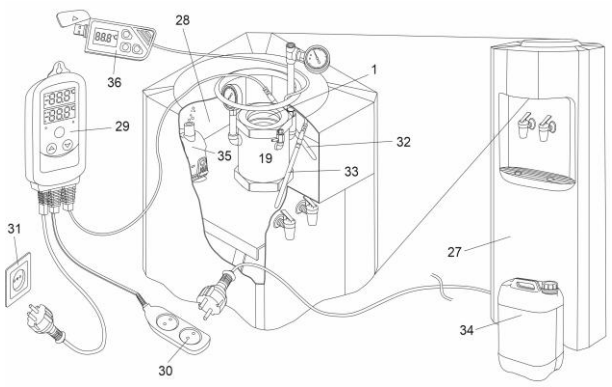

Figure 8. The chamber is immersed in an automatically regulated cold bath prepared in the internal tank of the floor-standing water dispenser: 1 – upper sight glass; 19 – chamber; 27 – floor-standing water dispenser; 28 – internal tank; 30 – temperature controller socket; 31 – wall socket; 32 – temperature controller sensor; 33 – temperature logger sensor; 34 – canister; 35 – submersible pump; 36 – temperature logger

A detailed description of the water dispenser's internal tank is shown in Figure 8. To convert the water dispenser's internal tank into a cold bath, the thermal switch must be disabled. Usually, the thermal switch maintains the water temperature inside the range suitable for drinking, but this range is not adequate for forming methane hydrate. After hot-wiring the thermal switch, the internal tank of the water dispenser can be accessed by turning it to the side and removing the bottle holder on its top. Temperature regulation within the range of -20 to 5°C is achieved by a temperature controller that, with its operating specifications, maintains the temperature within this range. The water dispenser cord is plugged into the temperature controller socket, while the temperature controller's cord is plugged into a wall socket. The temperature controller's sensor is placed inside the water dispenser's internal tank. The interior of the water dispenser's internal tank is filled with a prepared mixture of ethylene glycol and water from a canister. Ethylene glycol lowers the freezing point of water, allowing temperatures below 0°C to be reached. The temperature controller operates the dispenser's cooling system based on the difference between the set and sensed temperatures.

In this way, the temperature controller maintains the cold bath temperature at the set value. A submersible pump (e.g., aquarium type) enables the cold bath to circulate gently in the water dispenser's internal tank, ensuring a uniform temperature. The temperature logger records temperature changes using its sensor, which is also immersed in a cold bath. This type of temperature logger, in the form of a USB drive, allows recorded data to be transferred to a computer immediately after the methane hydrate formation process is complete. Once methane hydrate formation is visually confirmed through the upper sight glass, it is desirable to further lower the temperature before removing the chamber from the cold bath, so that the hydrate remains stable for as long as possible outside of conditions that favor it. Since the chamber is still under a certain pressure after the formation of methane hydrate, to safely remove the upper sight glass and access the methane hydrate, a bleed valve bar handle is passed through the rounded opening at the top, whereby the screw located inside the bleed valve is then unscrewed (Fig. 4). The screw inside the bleed valve is raised above its seat, while the chamber is depressurized in a controlled manner.

Since the kinetics of natural gas-hydrate formation within geologic materials is perhaps one of the less studied aspects related to their occurrence in nature, a new direction of the laboratory equipment development should be the creation of a chamber in which, at critical pressure and temperature values, it will be possible to simulate the lithological conditions of the soil (permafrost) or rocks (ocean floor) in which methane hydrates have been found so far [31]. Difficulties in investigating hydrate formation kinetics in porous media through laboratory experiments are mainly related to controlling heat and mass transfer within the porous medium [32]. However, this approach would enable the creation of highly accurate geological-numeric models for potential deposits. Since the hydrate-forming methane is either generated in situ or migrates upward from deeper sources, an apparatus capable of simulating methane transport processes is also required.

4. Conclusions

The conceptual model presented in this paper is a chamber for producing methane hydrates under laboratory conditions. It includes the use of accompanying equipment of spatially acceptable dimensions suitable for smaller experimental and working spaces. It is intended for use in university conditions, where research teams often have limited financial resources, and for which methane hydrates, as a future energy source, are of interest.

According to an internet search (e.g., Google Patent Search), it is possible to find solutions with designs different from ours that provide the same technical solution as our conceptual model. However, all these solutions still require a substantial financial investment. Explained in this way, the invention may also be refined and improved. For example, a submersible magnetic mechanism could enable the mixing of methane and water through a control glass at the bottom of the chamber, since the glass is a non-magnetic material and allows magnetic fields to pass through. However, the spirit of the invention is fully reflected in this paper with a well-defined basic principle of its operation. This conceptual model has also been submitted to State Agency for Intellectual Property in Zagreb, Croatia, for evaluation and is currently under review.

Author contributions

Conceptualization: MU, TM; Data curation: MU, LU; Formal analysis: MU, LU; Funding acquisition: TM; Investigation: MU, TM, LU, BP; Methodology: MU, TM; Project administration: TM; Software: LU, MU; Supervision: TM, BP; Visualization: MU, LU; Writing – original draft: MU, TM, LU, BP; Writing – review & editing: MU, TM, LU, BP. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interest

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in the study are included in the article; further inquiries can be directed to the corresponding author.

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Походження і значення метанових гідратів та їх утворення в лабораторних умовах

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Мета. Метою даної статті є представлення концептуального проєкту лабораторного обладнання для контрольованого утворення та дослідження малих кількостей метанових гідратів за змінних умов тиску, температури й літологічного середовища, що дозволяє проводити відтворювані експериментальні дослідження з імітацією природних умов формування гідратів метану.

Методика. Дослідження включає огляд сучасних світових наукових напрацювань і пілотних проєктів, пов'язаних із розвідкою, поширенням та видобуванням метанових гідратів. Узагальнено основні геологічні та термодинамічні умови, що визначають їх формування. На основі проведеного аналізу розроблено інженерну концепцію лабораторного апарата, яка передбачає використання камери високого тиску, системи стиснення газу та вузла температурного контролю, здатних працювати в межах зони стабільності метанових гідратів.

Результати. Запропонований апарат має компактну та модульну конструкцію і придатний для використання в стандартних умовах університетської лабораторії, що забезпечує контрольовану зміну параметрів тиску й температури, необхідних для утворення метанових гідратів, із забезпеченням безпеки експлуатації та відтворюваності процесу. Конструкція грунтується на використанні комерційно доступних компонентів і може бути реалізована з відносно помірними фінансовими та технічними витратами.

Наукова новизна. Представлене рішення є практичною та економічно доцільною альтернативою складним і дорогим лабораторним системам, які, зазвичай, застосовуються для дослідження метанових гідратів. Оригінальність підходу полягає в інтеграції доступних промислових компонентів у спрощену експериментальну установку, здатну відтворювати ключові умови формування метанових гідратів у контрольованому лабораторному середовищі.

Практична значимість. Запропонована лабораторна система надає дослідникам, викладачам і студентам гнучку експериментальну платформу для вивчення метанових гідратів у водному середовищі або пористих матеріалах. Вона сприяє проведенню міждисциплінарних досліджень у галузях геонаук, енергетичної інженерії та екологічних досліджень, а також може ефективно використовуватися в освітньому процесі та експериментальній науковій роботі.

Ключові слова: *метанові гідрати; зона стабільності газових гідратів; нетрадиційні газові колектори; лабораторне обладнання*

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