High-Pressure and Automatized System for Study of Natural Gas Hydrates

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Abstract: Due to the declining of oil reserves in the world in the coming decades, gas hydrate (GH) is seen as the great promise to supply the planet's energy demand. With this, the importance of studying the behavior of GH, several researchers have been developing different systems that allow greater truthfulness in relation to the conditions where GH is found in nature. This work describes a new system to simulate formation (precipitation) and dissociation of GH primarily at natural conditions at deep-sea, lakes, and permafrost, but also applied for artificial gas hydrates studies (pipelines, and transport of hydrocarbons, CO_2, and hydrogen). This system is fully automated and unique, allowing the simultaneous work in two independent reactors, built in Hastelloy C-22, with a capacity of 1 L and 10 L, facilitating rapid analyses when compared to higher-volume systems. The system can operate using different mixtures of gases (methane, ethane, propane, carbon dioxide, nitrogen, ammonia), high pressure (up to 200 bar) with high operating safety, temperature (-30 to $200\,°C$), pH controllers, stirring system, water and gas samplers, and hyphenated system with gas chromatograph (GC) to analyze the composition of the gases formed in the GH and was projected to possibility the visualizations of experiments (quartz windows).

Keywords: gas hydrate; energy; reactor

1. Introduction

Gas hydrate is a cage-like lattice of water molecules inside of which are trapped small molecules, such as CH_4, CO_2, H_2S, N_2, ammonia, H_2, and light hydrocarbons. Natural gas hydrates are constituted mainly by methane [1,2]. The expressive amount of GH around the world revealed an immense reservoir of carbon (0.5–12.7×10^{21} g) [3,4]. When brought to the earth's surface, one cubic meter of methane hydrate releases 164 cubic meters of CH_4 [5]. If converted to energy, this amount of methane is twice the amount of fossil resources already found and, therefore, consist of an opportunity for future exploration. Natural hydrate deposits may be several hundred meters thick and usually occur in two types of settings: Under Arctic permafrost and below the ocean floor in special conditions of temperature, pressure, and gas concentration [2].

The non-conventional resources will be an important alternative energy source at a worldwide scale to supply the planet's energy demand, because seismic data estimate that GH reserves are the largest sources of energy in the world. Nowadays, innovative research projects explore pathways to

obtain natural gas from gas hydrates (GH). Beyond this, gas hydrates have also been considered as a potential way to transport and store hydrocarbons, CO_2, and hydrogen [6].

Nowadays, many researchers have been developing systems to synthesize gas hydrates with different formation conditions and synthesis methodologies to understand the thermodynamics and kinetic of hydrate formation and dissociation. This knowledge is very important for gas hydrate exploration and exploitation techniques, but also for the transport of hydrocarbons in pipelines. It is also important to understand the dynamics of natural gas hydrates systems because their stability (both in permafrost regions and in deep-sea settings) will be affected by climate change [7]. Beyond this, researchers have been studying the application of different compounds as gas hydrate inhibitors (kinetic or thermodynamic), such as salts, amino acids, and alcohols. These inhibition experiments are usually performed in smaller reactors (e.g., 0.655 L) due to the higher precision in controlling the experiment conditions and gas consumption [8,9].

The first studies on the stability of GH occurred in early 1970s and the experiments were carried out in rustic reactors, without automation [10]. In the 1980s, automation started to be used (small technological advances) [11,12], and in the 1990s, some studies substantially advanced in reactors automation (control of temperature, pressure, and stirring) and in the use of new materials, such as acrylic or sapphire cells, but reactors sizes were still small (e.g., 300 mL) [13]. The main advantage of acrylic and sapphire reactors is the possibility to visualize the reaction during the experiment while the stainless-steel reactors have low cost. At this time, the researchers generally use only methane as host gas [11,12,14]. In the 21st century, several researchers published studies on GH synthesis, where they used sediment in 1 L and 10 L reactors with temperature control (thermocouples), pressure, and quartz or sapphire windows to allow visual inspection of the formation of hydrates, and using material with higher corrosion resistance (simulation of marine conditions) was used to synthesize methane, CO_2, and other gas hydrates [15–38]. Some other works started dealing with large-volume reactors (from 10 to 1710 L) trying to get closer to natural conditions [39–41].

This work describes a new system hereafter called Pilot Plant to study gas hydrates formation (precipitation) and dissociation primarily under natural conditions such as the ones found in deep-sea (or lake) and permafrost environments, but also the conditions found in pipelines and during transport of hydrocarbons, CO_2, and hydrogen as hydrate. It is important to mention that this system is unique around the world due to the accurate measurements of flow control of the gas mixture (it allows the injection of an exact amount of methane, ethane, propane, carbon dioxide, nitrogen, ammonia). In addition, the system controls pH, temperature, and pressure conditions in two different considerable reactors volume (1 L and 10 L reactors) to realize experiments with or without sediments. The fully automated system was also projected to allow visual observations of the reaction (quartz window) to accompany phase changes during the entire hydrate forming/dissociating processes. Beside this, it has a gas collection system for isotopic analysis, as well as being connected to the GC to determine the proportion of gases that really formed the GH. In order to validate and show the good functioning of the pilot plant, some experiments are presented and the results are in agreement with the literature.

2. Materials and Methods

2.1. General Pilot Plant Description

The pilot plant was built to synthesize gas hydrate using different gases, such as methane, ethane, propane, carbon dioxide, nitrogen, and ammonia, as well as a combination of all these gases, with the exception of the NH_3 and CO_2 mixture, as they would form the solid ammonium carbamate compound in the mixer and tubing. If the experiments need to inject more than one gas, there is a gas mixer that ensures the homogenization of mixtures before entering the reactors. The system is composed of two different volume reactors with maximum pressure of 200 bar. One of them has 1 L (reactor 1) and the other one has 10 L (reactor 2) capacity. Each reactor has two quartz windows (located 180°) to allow visual observation of gas hydrate formation during the experiment. The panels' controls (R02 and R01)

can ensure real-time monitoring of pH and temperature values during the entire experiment while the general control panel shows the individual reading of pressure inside each reactor. The peristaltic pumps provide the injection of liquefied gases (ethane, propane, CO_2, and/or NH_3) in the reactors, while the booster ensures the pressure of methane and/or nitrogen gas in the reactors, and the chiller ensures constant temperatures in the study of gas hydrate formation (Figure 1). The next topics will discuss each part of the pilot plant to provide a better understanding of the system.
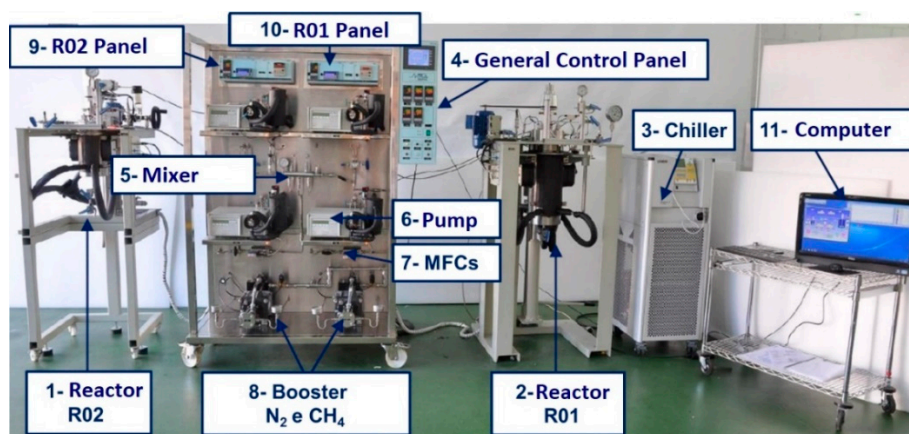
Figure 1. Main components of the pilot plant to study of gas hydrates: (1) Reactor R02 (1 L); (2) reactor R01 (10 L), (3) chiller, (4) general control panel, (5) gas mixer, (6) peristaltic pumps for liquefied gases (ethane, propane, CO_2, and/or NH_3), (7) mass flow controllers (MFCs), (8) booster for N_2 and CH_4, (9) R02 panel, (10) R01 panel, (11) controlling unit.

2.1.1. Characteristic of Reactors

According to the literature, most of the reactors are built by stainless steel [41–44], and some are made by acrylic [45], sapphire [46], and Hastelloy C-22 (a nickel-chromium-molybdenum-tungsten) [37]. The main advantage of acrylic and sapphire reactors is the possibility to visualize the reaction during the experiment while the stainless-steel reactors have low cost. However, the Hastelloy C-22 is highly resistant to corrosion and the material lasts longer. In this work, Hastelloy was chosen as the standard material for reactors and all wet parts of the pilot plant, mainly due to its resistance to corrosion, which allows simulation of marine environment (high salinity, low temperature, and high pressure) without damaging the equipment. The other parts of the pilot plant PPSGH were built of SS 316 L and all the components were obtained from Autoclave Engineers, Gyrolok, Swagelok, Classic Filters. Quartz windows allow visualization of reactors' interior.

In order to create a system that is representative of nature conditions and facilitate formation and dissolution of gas hydrate, the pilot plant here described uses two reactors with 1 L and 10 L, respectively, because the system requires small amounts of gas, does not require several days to put and remove the reaction, is easier to assemble and disassemble the experiments, requires lower investments when compared to the systems with higher volumes, is a flexible system with simultaneous performance of experiments with different reactional conditions (temperature, pressure, gaseous mixtures, etc.) and has the possibility of a reactional scale up (Figure 1). Besides that, it allows the use of sediments in the two reactors, because in systems of smaller volumes, it is very difficult to fit in cores and perform these experiments. As the only difference is the volume of the reactors, Figure 2 (10 L Reactor) displays some details of the system that are equivalent for both reactors.

Table 1 presents the main dimensions of the 1 and 10 L reactors, including the information about the two quartz windows located on both sides (180°) of each reactor. These windows (Figure 2) are a remarkable feature of the project, because they allow visualization of the hydrate formation while the experiment is being performed, providing an alternative way of confirming the presence of hydrate in addition to the recorded temperature and pressure data.

Table 1. Main design information about both reactors.

Data	Reactor R01	Reactor R02
Internal diameter (inch)	3.00	6.75
External diameter (inch)	6.19	8.38
Total height—with external parts (inch)	25.12	36.44
Inside height (inch)	8.87	17.38
Stirrer and thermowell height (inch)	7.68	16.34
Window center position from the bottom (inch)	6.87	12.75
Windows width (inch)	1.83	3.69
Window quartz space (inch)	0.50	0.50
Total volume (L)	1.0	9.5
Maximum pressure (bar)	221	221
Minimum temperature (°C)	−29	−29
Maximum temperature (°C)	177	177

Large-volume systems that can operate on high pressure conditions are considerably complex mainly due to material and security issues. The reactors in this work are capable of working until 20 MPa and to ensure the experiments are high pressure, the system contains busters able to increase the gas pressure to values as high as 20 MPa.

Two different proportions of gases form the GH in the most varied reactional conditions. The reactors are equipped with a gas purge system (Figure 2), which allows the venting of the gas inside the reactor with nitrogen after the hydrate formation. This technique can maintain the pressure when the hydrate was formed while purging the reactors headspace. After this purge, the system is closed and the user starts to increase the temperature or decrease the pressure slowly, causing the hydrate to dissociate. As the hydrate is decomposing, the sampling valve at the exit of the reactor is opened and the gas being released by the hydrate dissociation can be directly analyzed in the GC. This is a remarkable feature because, due to the different solubility of each gas in water, the hydrate composition can be different from the composition of the inlet gas or the gas phase after the hydrate formation and before the purge with nitrogen. In addition, to analyze the water phase, a valve located at the base of the reactor permits liquid sampling at any time during the experiments without depressurization of the system (Figure 2).
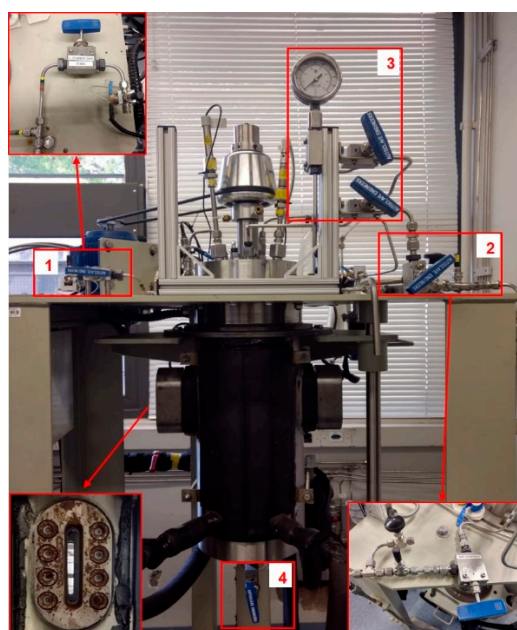
Figure 2. Illustration of the 10 L reactor showing the described features of the system. (1) First sampling point; (2) second sampling point; (3) purge system; and (4) water sampling point.

### 2.1.2. Controls Parameters: Temperature, Pressure, Stirring Speed, and pH

The pilot plant has a chiller (brand Lauda-XT 550) with an extremely versatile system, which is able to work between −40 °C and 200 °C, making it possible to perform a wide range of experiments. This way, it is possible to synthesize the gas hydrates in their stability zone in deep-sea conditions (nearly 0 °C to 19 °C), according to pressure, salinity, gas composition, etc.

The temperature in the reactor is measured by a Type T thermocouple housed inside a thermowell in each reactor (Figure 3). The thermocouple signal is assessed by the controller, which, depending on the difference with the set point, controls the opening of the electronic valves in the chiller line, and therefore, controlling the flow of coolant liquid (ethylene glycol/water) that goes through the cooling jackets of the reactor. Due to the size of the reactors, the stirring is an important factor for a homogeneous temperature profile inside the reactor and, therefore, a correct temperature control.

Each reactor shows a pressure control based on an E model Swagelok pressure transducer. These controls compare the set-point desired by the user to the actual pressure inside the reactors obtained from the transducers and act on the electronic valves to release a part of the gas or completely close/open the system.

In addition, the pilot plant system uses a blade stirrer from Magnedrive (Figure 3) for each one of the reactors, allowing a variation of 0–1300 rpm on the reaction system. The stirring speed during the experiment is very important to understand the influence of this factor on the hydrate formation process [42–44].

A Hastelloy C-22 pH probe (Corr Instruments) is responsible for real-time pH measurements during the experiments. It can work until 200 bar but it cannot be used at negative temperatures, as they damage the membranes that measure the concentration of H^+ in the reaction medium. The objective of a pair of pH electrodes (indicator and reference—Figure 3) is to check the pH variation during experiments. The pH probes are attached to the top of the reactor through a threaded connection of 1/4″ NPT (National Pipe Tapered) that seals this part of the structure not letting any gas leak. The pH probe height inside the reactor can be adjusted to be always inside the solution, regardless of its volume.
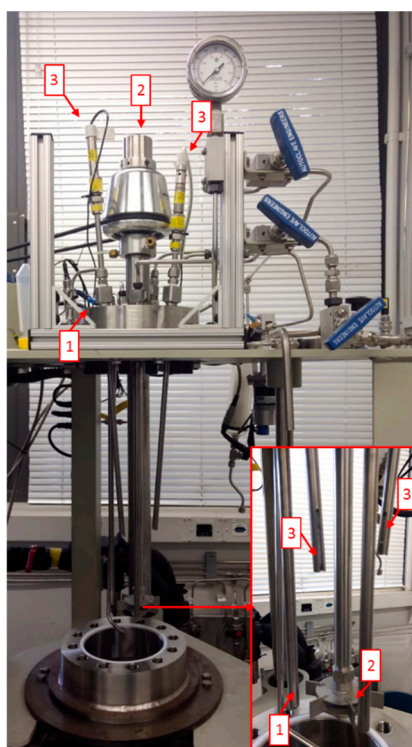
Figure 3. Internal of the 10 L reactor showing the described features of the system. (1) Type T thermocouple; (2) stirrer; and (3) pH probes.

2.1.3. Gases System

According to the literature, gas hydrates are formed mainly from light hydrocarbons (e.g., methane and ethane), CO_2, and H_2S [2]. Therefore, a system with six different components (methane, ethane, propane, carbon dioxide, ammonia, and nitrogen) was designed to produce gas hydrates with many possible compositions.

Four of the six compounds are liquids at its vapor pressure: Ammonia, ethane, propane, and carbon dioxide. For this reason, high-performance liquid chromatography (HPLC) pumps are properly cooled to ensure the liquid injection in the reactors. The $NH_{3(líq.)}$, $C_2H_{6(líq.)}$, $C_3H_{8(líq.)}$, and $CO_{2(líq.)}$ cylinders (siphon type), must have connections to liquid outlet of the 6 bars, 33 bars, 6.5 bars, and 60 bars, respectively, at 15 °C. Liquefied gas cylinders do not require a pressure regulator. Instead, connect the line to the liquid outlet port and a pressure indicator.

The pilot plant has four high-pressure HPLC Gilson 307 pumps (Figure 4) with a maximum flow capacity of 5 mL/min (CO_2 and C_3H_8) and 10 mL/min (NH_3 e C_2H_6), respectively. It is important to highlight that the ammonia pump has a special seal of PTFE/graphite because of the high corrosion capacity of this compound. The maximum pressure for these HPLC is 600 bar. Each pump has a pressure transducer inside that allows it to set up alarms, reduce oscillation, and ensure a constant flow.

The cooling liquid from the pilot plant chiller is kept in a reservoir attached to the peristaltic pumps, from which it cools the head of the pumps to keep it at low temperatures and ensure there is no vapor formation inside the pump system. A main temperature controller set is used to check all the pumps' temperatures. These temperatures have to be kept at values lower than 0 °C so that the pump works properly, and cavitation is avoided.

Each pump has a manifold (Autoclave Engineers) fixed on the top of the pump that allows the user to purge the liquefied gas line before starting an experiment. Each pump has a back-pressure regulator that is responsible for creating an overpressure of 20 bar downstream of the pump, avoiding any liquid or gas to return and damage the pump.
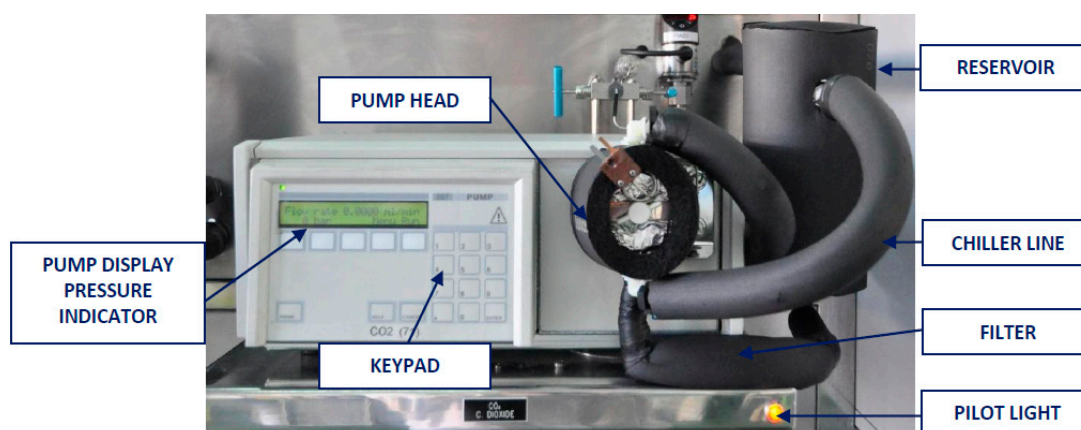

Figure 4. Carbon dioxide HPLC pump; one of the four pumps used to inject liquids in the reactors.

The other two compounds (N_2 and CH_4) are gases even in high pressures. Even so, two boosters (Maximator) were implemented to inject these gases separately and at each desired pressure (Figure 5). The gas flows are controlled by two mass flow controllers (MFC), which also show the amount of gas being injected in mL/min. Since they are mass controllers, gas temperature and pressure does not influence on the measurement. Since boosters are air driven, they require a pneumatic air facility, of 12 mm diameter, capable of supplying approximately 2000 N/min of air at 8–10 bar.
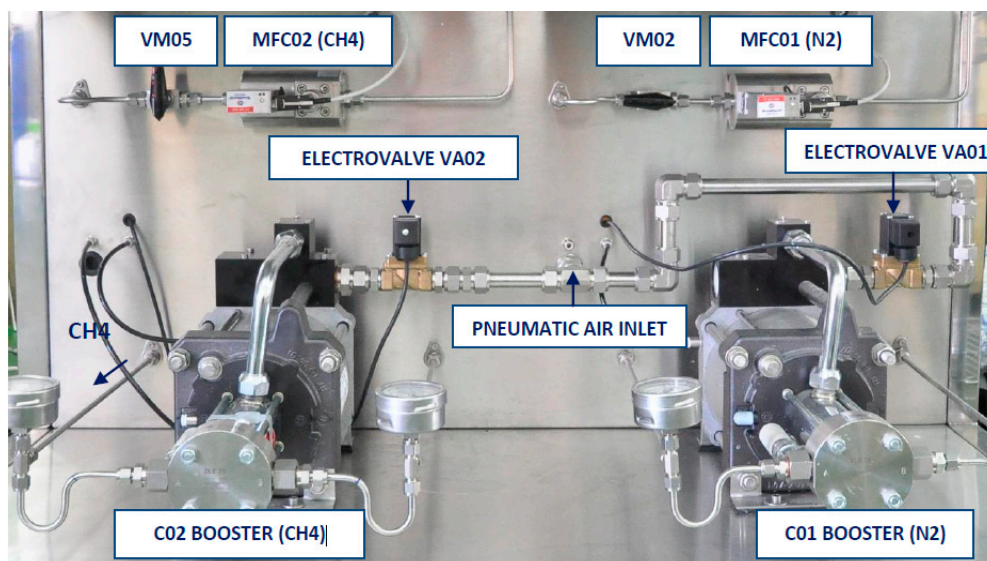
Figure 5. Methane and nitrogen boosters. VM05 and VM02—globe valves; MFC01 and MFC02—mass flow controllers; VA01 and VA02—electrovalve for inlet air control for the booster.

Besides this, two vessels of 2.5 L each (Figure 6) are located on the back part of the plant and are responsible for reducing the pressure oscillation after the thrusters and inject a continuous flow of gas. After being thrusted, the gas is kept under a controlled pressure inside the vessels. This pressure must be higher than the reactors pressure (at least 30 bar) so that the gas can be sent to reactor through the MFC. Inside the reactor, the pressure is controlled based on two other instruments, depending on which reactor is being used. Each vessel has a vent valve, a rupture disk, and a pressure transducer used for pressure control. Due to the separate control systems, both gas pressure and reaction pressure can be set at different values, making the system more flexible.
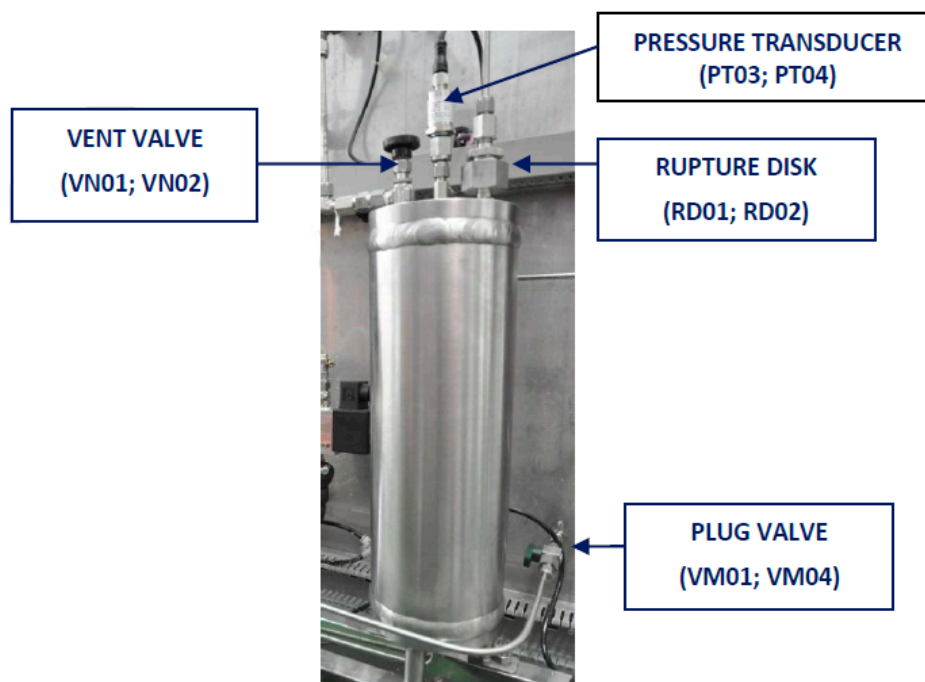

Figure 6. Vessels used to storage the gases (N_2 and CH_4) before entering the reactor. VN01 and VN02—storage vessels vent valve; PT03 and PT04—pressure transducer for pressure control; RD01 and RD02—safety rupture disk; VM01 and VM04—plug valve for gas inlet.

2.1.4. Gases Mixer System

The possibility of using different gases at the same time or a gas mixture with a wide range of composition is one of the main advantages of this system, leading to a versatile operation with countless opportunities of experiments. To do so, the system has a mixer (Figure 7) that consists of a SS316 element in which five of the reactants (CH_4, NH_3, C_2H_6, C_3H_8, and CO_2) can be mixed before entering the reactors. A check valve in each line prevents backflow of any of the components. A manometer at the outlet shows the pressure of the mixer and it is also equipped with a rupture disk and a vent valve for security reasons.

Upon leaving the mixer, the compounds are driven to the main manifold (VM10), which allows for selecting one of the two reactors (R01—right, R02—left). It is important to highlight that the NH_3 and CO_2 should not be mixed in this system. These two elements together react and produce a solid salt known as urea ($CO(NH_2)_2$) that can plug the system and damage its operation. It is possible to mix N_2 with the rest of the gases by opening the valve VM11, which connects the N_2 MFC to the mixer, making sure that the pressure in the N_2 vessel is higher than the pressure in the mixer (Figure 7). It is also equipped with a check valve to prevent backflow of any of the compounds towards the N_2 line.
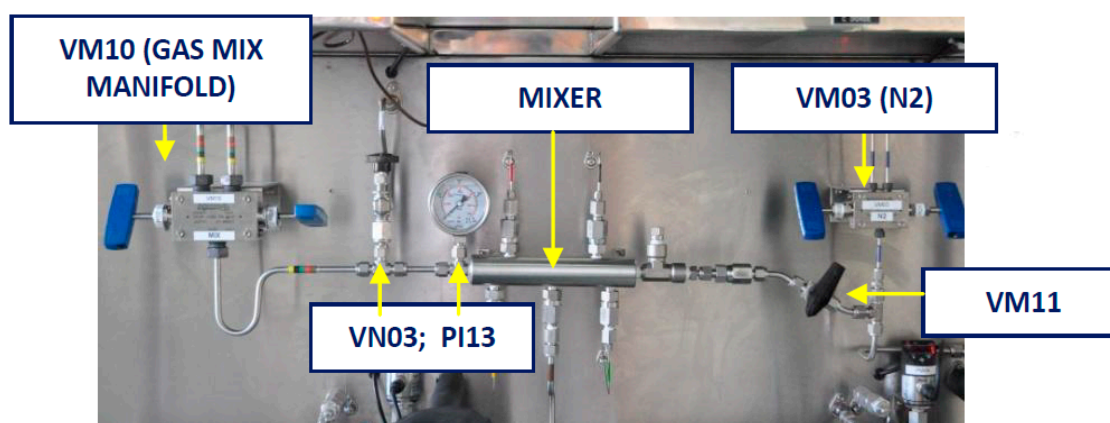
Figure 7. Gas and liquid mixer system. VM10—gas mixture manifold for gas feed to the reactors; VN03—mixer vent valve; PI13—mixer manometer; VM03—nitrogen manifold for pure nitrogen feed to the reactor; VM11—nitrogen valve that connects the nitrogen line with the mixer.

Pressurizing the mixer with N_2 prior to pumping liquefied gases is advisable. Otherwise, the pressure must be reached using liquefied gases, which would take much longer than pressurizing with a gas. Both reactor inlets are protected with check valves. For this reason, when the reactor is being pressurized at the beginning of the experiment, the manometer in the mixer will show lower pressure than the reactor. Pressurization of mixer with N_2 is possible by valve VM11. Besides that, N_2 can also be introduced into the reactors independently from the rest of the gases, thanks to the manifold (VM03). With this feature, it is possible to perform leaking tests in the system using nitrogen.

2.1.5. Gases Injection System

The pilot plant has six Swagelok flow control valves that control the compounds flow for each part of the system. These valves are operated through the control system of the pilot plant, allowing the user to input a set-point of desired flow of each gas or liquid that will be sent to the mixer or directly to the reactor. This allows the user to record the amount of gas used in every experiment, in any reactor. After the six flow control valves of each reactant, there is the flow control valve located between the mixer and the reactors, which allows for the user to know not only the amount of each compound, but the flow of mixture being injected in the system.

Besides the flow control, the system has four pneumatically actuated shut-off valves. The function of these diaphragm valves is to shut off the flow of liquefied gases and prevent the emptying of the

supply cylinders when the plant is not working. The liquefied gas cylinders do not work with pressure regulators at liquid outlet and, therefore, if the cylinder pressure is higher than the pressure in the pilot plant, the liquefied gas will flow freely if there is no restriction. As an additional safety measure, a manual 1/4″ Swagelok shut-off valve has been fixed before each diaphragm valve.

Another important feature of this plant is the fact that it can be fully pressurized with gas, N_2, and/or CH_4. This way, when liquids such as CO_2 are injected directly in the solution, the atmosphere inside the reactor is already under pressure and filled with the desired gas. This makes it possible to produce CO_2 hydrate only using liquid CO_2, for example, due to the fact the initial pressure necessary for the formation, close to 40 bar, can be reached with N_2 pressurization.

2.1.6. Panel Control System

The plant is controlled by means of a main control unit and two independent units for each reactor. The pressure control is centralized in the main control unit, as well as the alarms (Figure 8). Thus, the temperature and stirring of each reactor is set in their own control units. Before starting the experiment, the user selects which reactor will be used. The control system will assess if the conditions in the selected reactor are accomplished in order to allow for running an experiment. This security protocol certifies that the experiment conditions are inside the security limits and reduces the chances of human mistake. The main two factors to be analyzed before the experiment runs are pressure and temperature.
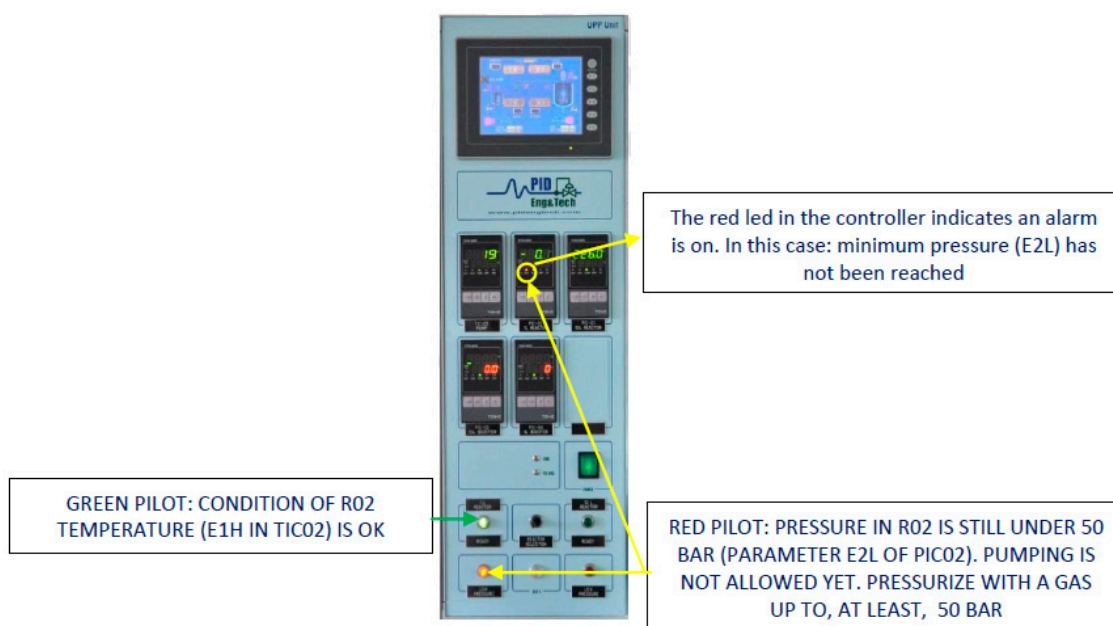
Figure 8. Main control unity showing the main alarm and indication lights.

2.1.7. Software for Plant Control

The pilot plant is equipped with a distributed control system with independent control loops for each variable. All the PID controllers are configurable and can be connected to alarms in the main control system. The digital communication between the controllers and a computer is done with the system called SCADA (Process@software, developed by PID Eng & Tech). The system can be controlled either from the main unity or from the computer. Besides that, the software used by the system allows the user to perform experiments step-by-step with everything being pre-programed and it is able to acquire and record all the measured variable from the experiment. Figure 9 shows the interface of the software used in the pilot plant.
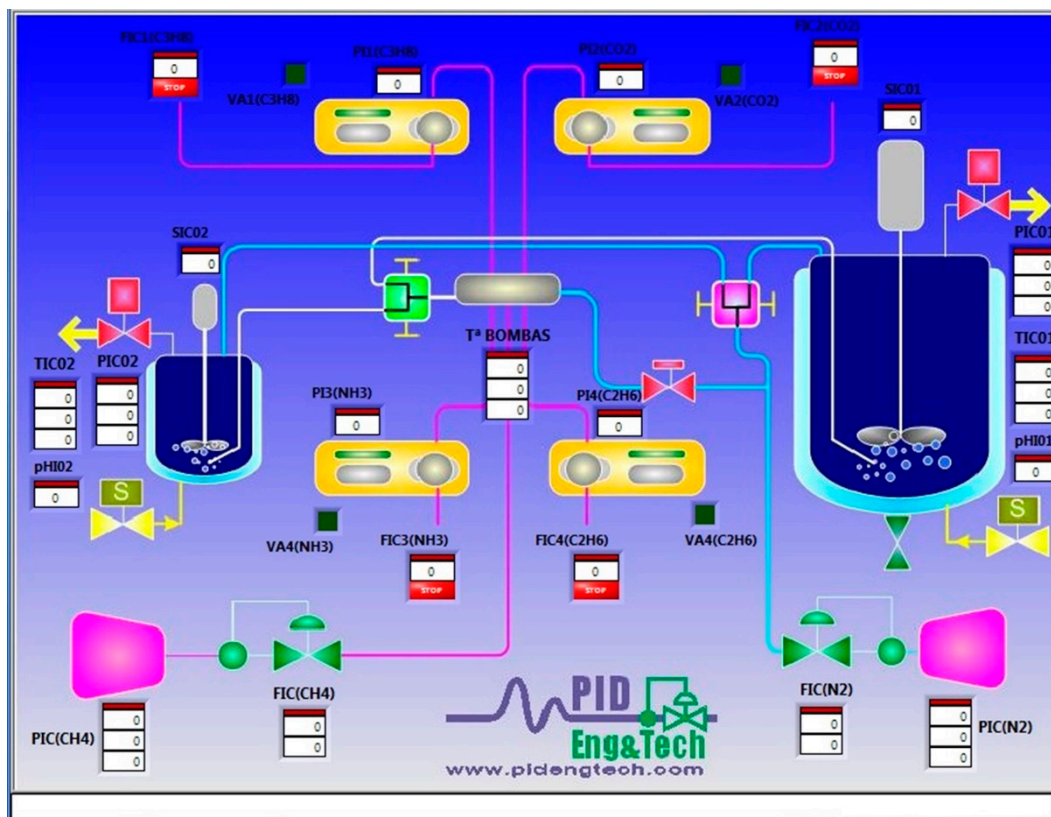
Figure 9. Pilot plant software interface (PID Eng & Tech).

3. Experiments Validation for Gas Hydrate Formation

In order to demonstrate and validate the good functioning of the pilot plant system, several experiments were performed. Among these, four experiments of GH synthesis simulating natural condition were done to obtain experimental data of carbon dioxide and methane hydrates in pure (Figure 10a,b) and saline (Figure 10c,d) water. The experiments were conducted in batch reactor mode under isochoric condition. The CO_2 and CH_4 hydrates were obtained simultaneously under the same reaction conditions in 1 L and 10 L reactors, respectively, proving that the system allows for the performing of two experiments with different gas composition at the same time, both resulting in hydrate formation. Firstly, 350 and 7000 mL of deionized or saline water were added in the 1 L and 10 L reactors, respectively, and then vacuum was performed for 6 h prior to the introduction of the respective gas in order to remove the air contained inside the reactor.

After that, the reactor was pressurized up to 40 bars of CO_2 at 12 °C. The temperature was set to decrease to 2 °C and when the system reached 6 °C, stirring was started at 300 rpm [9]. The temperature and stirring (2 °C and 300 rpm) were maintained until the formation of CO_2 hydrate. The synthesis of CH_4 hydrate were performed following the same steps, but the reactor was initially pressurized with 70 bars of CH_4. Figure 10a,c and Figure 10b,d show the graphs for carbon dioxide and methane experiments, respectively. Besides the described parameters, pH was also measured throughout the experiments. In the synthesis using CO_2 in pure water, pH decreased from 7.6 to 6.5 due to solubilization of CO_2 in water and consequently formation of H_2CO_3, while in the experiments using CO_2 in saline water, the pH decreased from 7.6 to 5.3, observing a greater solubilization of CO_2 in water and a greater formation of H_2CO_3. Regarding the CH_4 experiments, the pH remained almost constant, approximately 7.4 in both syntheses.
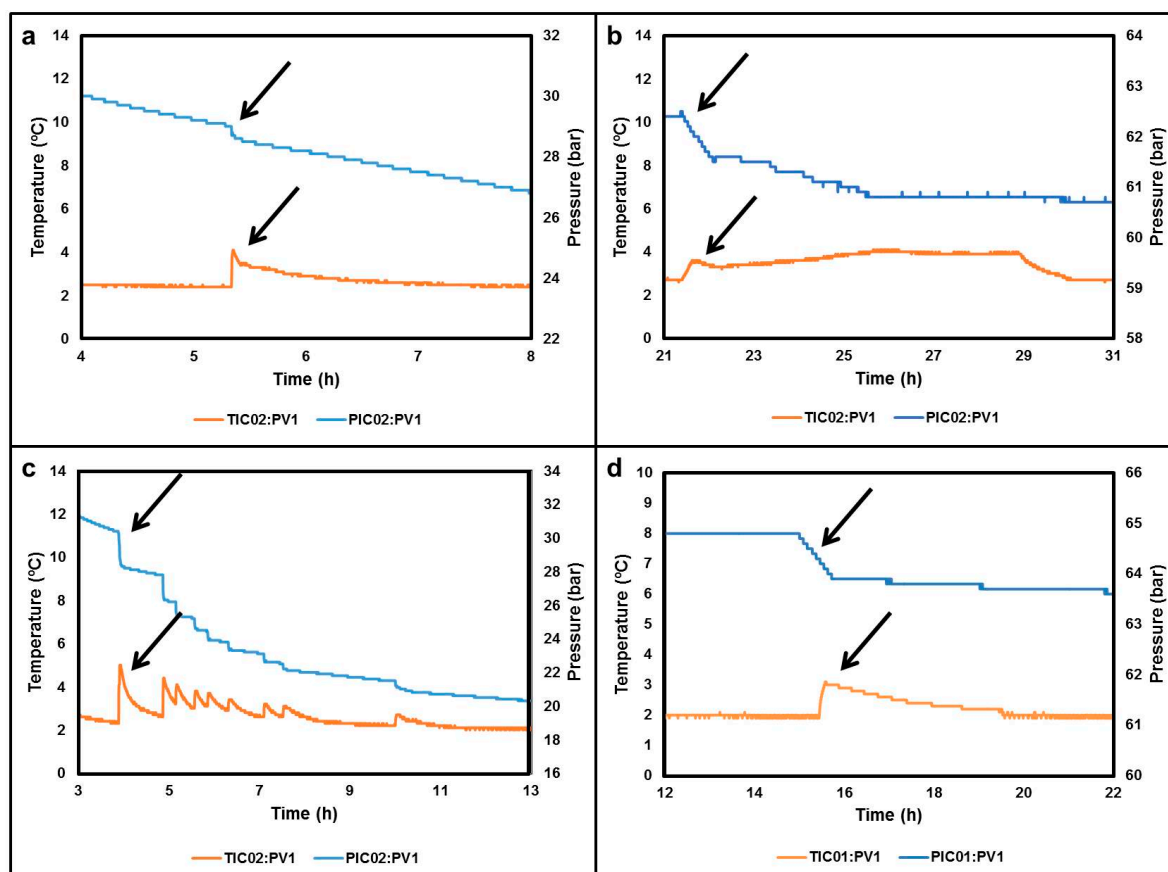
Figure 10. Gas hydrate (GH) synthesis with CO_2 in pure water (**a**), CH_4 in pure water (**b**), CO_2 in saline water (**c**), and CH_4 in saline water (**d**).

The hydrates formations were observed visually through the window present in the reactors, together with the pressure and temperature changes recorded by the Process software system, which makes the acquisition of data of all the variables involved during the experimental process in a time interval of 10 s [45]. The formation of GH is an exothermic process, which can be seen by the peak temperature at the beginning of the growth period as well as the pressure drop shown by the arrow, according to the illustration of Figure 10. The orange line, TICO2:PV1, corresponds to temperature and the blue line, PIC02:PV1, corresponds to system pressure. The black arrow shows the moment when the temperature increased followed by a decrease in the pressure. Throughout the process, pressure and temperature changes were monitored and recorded by the data acquisition system in a time interval of 1 s. Besides that, the hydrates' formations were observed visually through the window according to Figure 11.

According to the results presented, and compared with the literature [46], they reported on the balance of gas–hydrate–water phases as a function of temperature and depth. The main factor controlling the thermodynamic stability and spatial distribution of gas hydrates is the variation of methane solubility in water when compared to carbon dioxide. CH_4 is hydrophobic and interacts weakly with water, whereas CO_2 has a quadrupole moment leading to a relatively stronger interaction with water than CH_4 [47]. Thus, a greater time of formation of the CH_4 hydrate in relation to the CO_2 hydrate is observed, as can be observed in Figure 10.
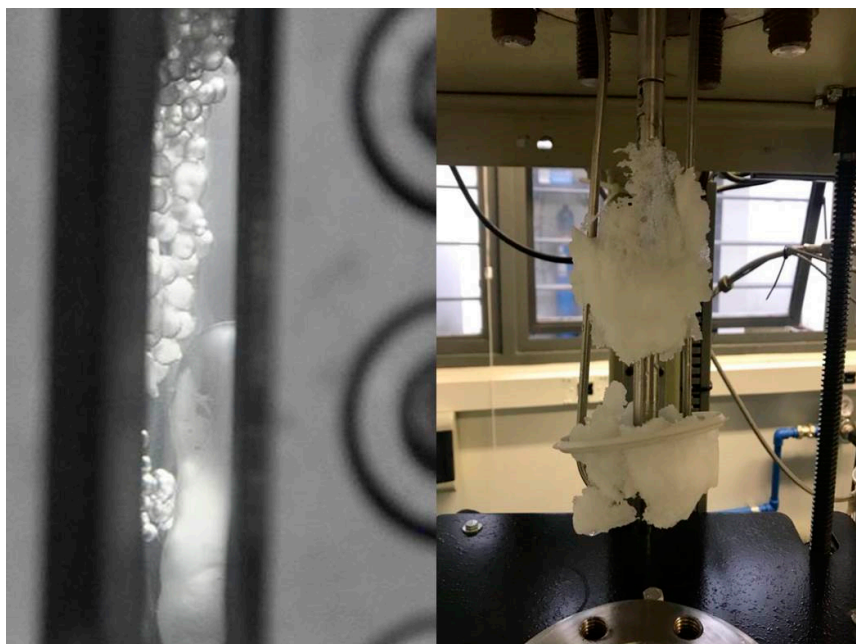
Figure 11. Images of the beginning of the formation of GH by the window and after the opening of the reactor.

4. Conclusions

This work described a new system (pilot plant) to study gas hydrates' formation and dissociation in permafrost, pipelines, and ocean conditions. A pilot plant has a sampling system to ensure the collection of liquid and gases and online monitoring during the experiments. Accurate measurements of flow control of the gases, pH, temperature, and pressure conditions were projected to monitor the phase changes during the entire hydrate forming process. In order to verify and validate the operation of the pilot plant, four experiments are described, using different gases to form, simultaneously, different GH, and the results are in agreement with the data contained in the literature.

The great novelties of this system in relation to existing systems are: (1) Possibility of injecting different gas mixtures without having to purchase them separately, which become any project more expensive; (2) possibility of conducting simultaneous and independent experiments using the same system; (3) work at different scales and, with this, visualize the differences in GH formation/dissociation; (4) ease of synthesizing HG in sediments when compared to very small or very large systems; (5) ease of cooling the system when compared to experiments on huge scales (72 to 1710 L); (6) low cost for system maintenance when compared to very simple systems and very low maintenance cost when compared to very large systems; (7) important for development of non-conventional energy exploitation.

Author Contributions: L.F.R., M.K., E.S. and R.L. were the creators of the pilot plant system for gas hydrate. In addition, M.K. was responsible for the project financed by Petrobras. A.R. and G.d.A. are the users of the system, since they performed the various experiments, proving the effectiveness of the pilot plant in the synthesis of gas hydrates with different compositions.

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