Potential Industry Applications Using Gas Hydrate Technology

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Abstract: *Over the past decades, gas hydrates have stimulated significant interest and triggered fundamental research. Primarily, the focus has been on hydrate blockage in pipelines, and on naturally occurring gas hydrates. However, gas hydrates can be useful in many different ways that can be pertinent to our industry, thanks to their unique structural packing where only certain molecules can enter the gas hydrate cavities. Among the several potential uses of gas hydrate technology are gas separation, transportation and storage of natural gas, desalination, and carbon dioxide disposal. In particular, it is possible to (i) separate the heavier components (pentane and higher) from natural gas, and (ii) capture, store and transport natural gas. This paper proposes a workflow for capturing, storing and transporting gas in the hydrate form, particularly for situations where there are infrastructural constraints such as lack of pipelines. These applications of gas hydrate technology can have potential benefits to the oil and gas industry.*

Keywords: *Natural Gas Capture Storage Transportation Hydrate*

1. Introduction

Hydrates consist of geometric lattices of water molecules containing cavities occupied by light hydrocarbons and other types of gaseous components such as nitrogen, carbon dioxide, and hydrogen sulfide. Although gas hydrates resemble ice or wet snow, in appearance, but do not have ice's solid structure, are much less dense and exhibit properties that are generally associated with chemical compounds. Gas hydrates of interest to the natural gas industry are made up of lattices containing water molecules in different ratios with methane, nitrogen, ethane, propane, iso-butane, normal butane, carbon dioxide and hydrogen sulfide. The cavities are at least partially occupied by small gas molecules such as CH_4, C_2H_6, C_3H_8, $I-C_4H_{10}$, $n-C_4H_{10}$, N_2, and CO_2 to stabilise the lattice structure.

The need for new methods for gas transportation is the challenge that drives the development of hydrate technology for storing and transporting natural gas (Masoudi, et al., 2005). The ability of natural gas to form hydrate in combination with water is a very interesting and useful concept (Makogon, 1997) and can be widely utilised in the industry. An important feature of hydrates is their high storage capacity. 180 volume units of gas at standard conditions can potentially be packed into 1 volume unit gas hydrate crystals (Sloan, 1997). Gas hydrates can be regarded as a safe and easy way of capturing gas, storing and transporting associated, stranded and flared gas (Berner et al., 2003).

The objective of this work is to propose useful industrial applications that rely on gas hydrate technology, based on selected gas samples (see Table 1). These applications include situations when:

1) Gas storage is required, and so natural gas is converted to gas hydrate and stored for future use.
2) Natural gas hydrate technology provides an attractive method to capture and transport natural gas on a small scale.
3) In the hydrate process of capturing natural gas, heavy components (C_5 and above) are separated out as Natural Gas Liquids (NGL), while C_1 to C_4 are stored in hydrate form.

Table 1. Natural Composition of the gas samples used (Mole %)

	N_2	H_2S	CO_2	C_1	C_2	C_3
Dry Gas	0.00	0.00	0.00	99.00	1.00	0.00
Sample 2	0.78	0.00	2.84	92.04	2.82	0.74

	iC_4	nC_4	iC_5	nC_5	C_6
Dry Gas	0.00	0.00	0.00	0.00	0.00
Sample 2	0.14	0.21	0.10	0.08	0.25

The proposed workflow will be discussed using two representative gas samples ('Dry gas' and 'Sample 2').

The former is basically pure methane, whereas sample 2 has the heavier C_5 and C_6 components. It is assumed that these gases are produced from a given field, at a given rate.

Most of the previous studies focused on simple gases with composition of primarily methane and ethane to form gas hydrate. Most natural gas has much more components than just methane and ethane and hence the composition can have significant impact on hydrate formation. For this reason, this analysis showed a dry gas sample and sample 2 with heavier components. The overall study looked at over 20 natural gas samples with varying composition.

In this study, the PVTSim program (Calsep, 2008) was used for the evaluation of hydrate formation and expansion processes. Expansion of the gas from wellhead conditions is necessary to trigger hydrate formation, depending on the properties of the gas, as it will be shown in this analysis with the two selected samples. Hydrate formation conditions of 600 psia and 35°F are assumed, based on laboratory studies conducted by Okutani et al., (2007) who used methane, which is close to the dry gas sample used in the present work.

In the hydrate formation process, only the C_1 to C_4 alkane components of natural gas are captured. C_5 and higher components are separated out as natural gas liquids. This is a particularly useful concept, especially in cases when the lighter components of natural gas are needed for power generation, and the heavier components can have a negative impact on gas turbines. This concept is illustrated with the Sample 2 case.

The formation of natural gas hydrate yields a high latent heat of formation that must be removed to prevent dissociation. To this aim, the formation vessel could be equipped with heat exchange tubes, extending the full length of the vessel, to facilitate heat transfer from the vessel. The heat exchange tubes not only aid the heat removal process - they also (i) supply heat for later dissociation of hydrate, after formation and storage/transportation, and (ii) provide additional surface area for more effective hydrate formation.

The focus of the study is to evaluate forming gas hydrate as soon as the gas comes from the well. Whether it is offshore or onshore the hydrate vessels will be position there to capture the natural gas from the well. Other studies focused on gas being transported to a hydrate plant for hydrate conversion and therefore the conditions using gas directly from the well in this analysis are different from other evaluations. Therefore, it is proposed that the same vessel used to form the hydrate be also used for storage and transport to its delivery point.

The "one vessel" concept is very useful to avoid moving the solid hydrate from vessel to vessel for storage and transportation, reduce costs, since no additional facility is needed for dissociation at the final destination, and allow water re-cycling. Using one vessel for formation, storage, transportation and dissociation of the hydrates gives operational flexibility for temporary storage and transportation. In the absence of pipeline infrastructure, hydrates could be transported in the vessel, by truck, railway or ship.

2. Dry Gas Sample Analysis

The process flow in Figure 1 illustrates the capture of 5 MMSCF of dry gas from one producing well in hydrate form. The wellhead conditions are considered to be 1,750 psia and 168 °F. From the wellhead, the gas flows through a turbo-expander, which causes the gas temperature to drop to 35 °F, and the pressure to drop to 600 psia, assuming an efficiency of 85% (note that some commercial expanders can exhibit up to 90% efficiency). These new pressure and temperature values represent the inlet conditions to the hydrate reactor vessel. Note the heating value of the dry gas is the same before and after hydrate formation (1,018 btu/ft^3).
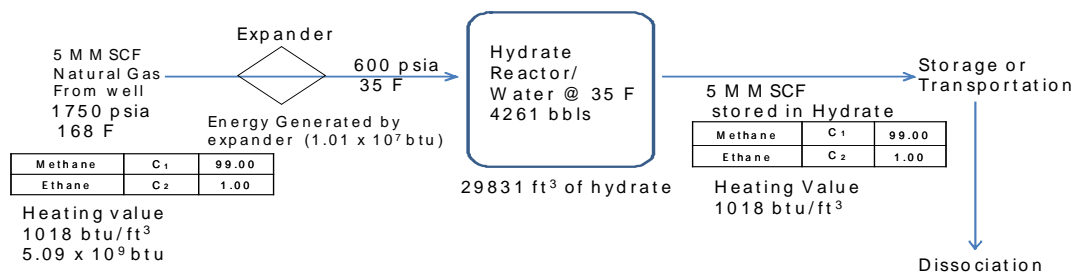

Figure 1. Gas Hydrate Process Flow for the Dry Gas Sample

The sample's heating value was estimated from the heating values of the sample's components, using the composition shown in Table 2. The amount of water required for the process was estimated at 6.29:1 mole ratio of water to gas for the Dry gas sample. This was determined from a sensitivity analysis using several samples with varying composition, and discussed in a previous paper (SPE 131663). A total of 4,261 bbls of water is therefore required to capture the 5MMSCF of gas.

Table 2. Heating Value Estimation for Dry Gas Sample

		Ideal Heating Value	Heating Value Gas mix in Hydrate	Heating Value Gas mix before hydrate
Sample Dry gas	Mol %	btu/ft3	btu/ft3	btu/ft3
Methane	C_1	99.00	1010.0	999.9	999.9
Ethane	C_2	1.00	1769.7	17.7	17.7
Total		100		1018	1018

Figure 2 shows the expansion process with corresponding energy exchange for the ideal process (isentropic and 100% efficient) and for the actual process at various expander efficiencies. The secondary axis of the graph is the outlet temperature that corresponds to a given efficiency. The figure presents the variation in enthalpy and entropy for the expansion process considering several expansion efficiencies. At 100% efficiency (Isentropic process), entropy is constant but the value increases as efficiency decreases. The work the gas performs is gained from its enthalpy and the gas cools rapidly in the expander. The expansion process must also ensure the gas remains in the gaseous phase.
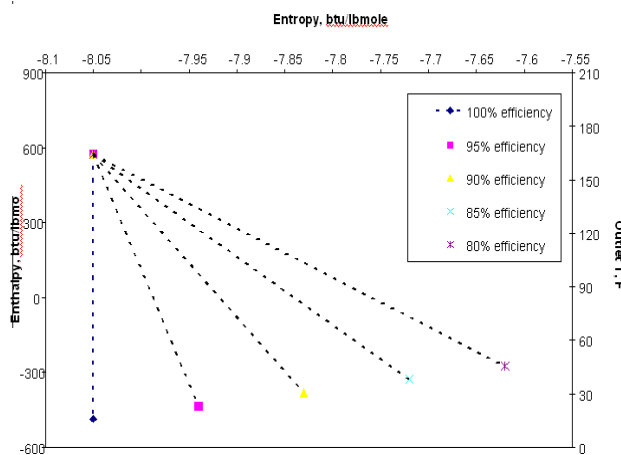

Figure 2. Expansion Process for Dry Gas Sample

The power developed by the expander and the outlet temperatures are shown in Table 3. An efficiency of at least 85% is required to obtain an outlet temperature of 35 F, required for hydrate formation. This generates 1.01 x 10^7 Btu of energy. Below is a sample calculation of power from the expander:

$$Power_{expander} = \Delta h \times w \times \eta_e$$

Where:

Δh = change in enthalpy, btu/lbmole, obtained from Figure 2.

w = flow rate, lbmole/hr

η_e = expander efficiency, %

For an 85% efficiency, Δh = 903.1 btu/lbmole and w = 550 lbmole/hr

$$Power_{expander} = 422199.3 \text{ Btu/hr}$$

Some commercial expanders can have up to 90% expansion efficiency, which is adequate for both samples to process 5MMSCF/D with only one expansion stage.

Figure 3 shows the phase diagram for Dry Gas sample, with the wellhead and outlet conditions for varying expansion efficiencies. It can be seen that gas remains in the gas phase region during the expansion process. Note that this sample is mainly pure methane and does not exhibit a phase envelope.

Table 3. Horse Power Generated and Outlet Temperature for various expansion efficiencies

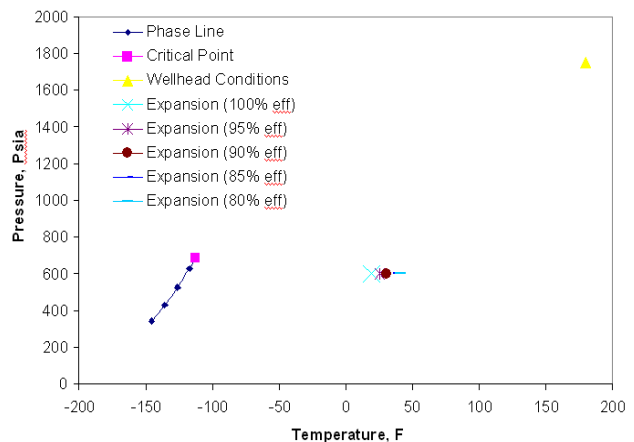| Efficiency	Power	Outlet Temperature
%	btu/h	
100	584375.0	19.4
95	527359.3	25
90	473319.0	30.3
85	422199.3	35.5
80	374000.0	41.3

Figure 3. Phase Diagram for the Dry Gas Sample

3. Sample 2 Analysis

The process flow in Figure 4 illustrates the capture of 5 MMSCF of Sample 2 gas in hydrate form. The computed heating value of the gas was 1,029 btu/ft^3 before the hydrate formation, and 1,014 btu/ft^3 after. This is because the hydrate formation separates the heavier components (> C5) as useful natural gas liquid. 0.02 MMSCF (3,829 bbls) of natural gas liquids are obtained with a heating value of 4,441 btu/ft^3 while 4.98 MMSCF natural gas (C_1 to C_4) is stored in hydrate form.

The estimations of the heating values of the gas prior to hydrate formation and the heating values transported in hydrate form for both samples were important to determine gas acceptance at markets around the world or whether further gas processing required (see Table 4). The heating value was calculated simply by

multiplying the ideal heating values of the individual component by the mole fraction which gives a simply approximation. This may vary from measured values.
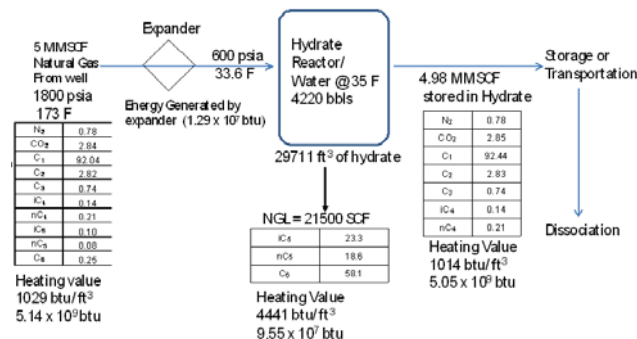

Figure 4. Gas Hydrate Process Flow for Sample 2

Table 4. Estimation of Heating Value of Sample 2 before hydrate formation

Sample 2 before hydrate formation		Ideal Heating Value	Gas mix before hydrate
		Mol %	btu/ft3	btu/ft3
Nitrogen	N$_2$	0.78	0.0	0.0
Carbon Dioxide	CO$_2$	2.84	0.0	0.0
Methane	C$_1$	92.04	1010.0	929.6
Ethane	C$_2$	2.82	1769.7	49.9
Propane	C$_3$	0.74	2516.1	18.6
Isobutane	iC$_4$	0.14	3251.9	4.6
N-Butane	nC$_4$	0.21	3262.3	6.9
Isopentane	iC$_5$	0.1	4000.9	4.0
N-Pentane	nC$_5$	0.08	4008.9	3.2
Hexanes	C$_6$	0.25	4755.9	11.9
Total		100		1029

Table 5 estimates the heating value of the gas before hydrate formation. Table 5 estimates the heating value of the gas captured in the hydrate, and Table 6 estimates the heating value of the Natural Gas liquids that is not captured in the hydrate. The accepted heating value range accepted in the US is in the range 966 – 1,120 Btu/scf. For Europe, the range is 940–1204 Btu/scf and for Japan 1,065 - 1,160 Btu/scf is required.

The amount of water required for the process was estimated at 6.264:1 mole ratio of water to gas. A total of 4220 bbls of water is therefore required to capture 4.98MMSCF of gas.

The actual wellhead conditions in this case are 1,800 psia and 173°F which is slightly different from the Dry Gas Sample. The expansion turbine extracts the potential heat energy from the gas, causing it to cool drastically from 173°F to 35°F.

Figure 5 shows the expansion process with corresponding energy exchange for the ideal and the actual processes. The power developed by the expander and the outlet temperatures are shown in Table 7. At least 90% efficiency is required to have an outlet temperature of 35°F required for hydrate formation in this case. This generates 1.29 x 107 Btu of useful energy.

Table 5. Estimation of Heating Value of Sample 2 after hydrate formation

Sample 2 after hydrate formation		Ideal Heating Value	Gas mix in hydrate
		Mol %	btu/ft3	btu/ft3
Nitrogen	N$_2$	0.78	0.0	0.0
Carbon Dioxide	CO$_2$	2.85	0.0	0.0
Methane	C$_1$	92.44	1010.0	929.6
Ethane	C$_2$	2.83	1769.7	49.9
Propane	C$_3$	0.74	2516.1	18.6
Isobutane	iC$_4$	0.14	3251.9	4.6
N-Butane	nC$_4$	0.21	3262.3	6.9
Total		100	4000.9	4.0

Table 6. Estimation of Heating Value of Natural Gas Liquids

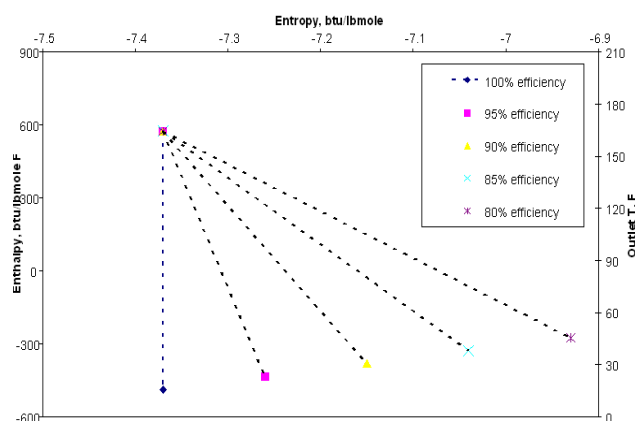| Natural Gas Liquids		Ideal Heating Value	NGL Heating Value
		Mol %	btu/ft3	btu/ft3
Isopentane	iC$_5$	23.3	0.0	0.0
N-Pentane	nC$_5$	18.6	0.0	0.0
Hexanes	C$_6$	58.1	1010.0	929.6
Total		100.0	1769.7	49.9

Figure 5. Expansion Process for Dry Gas Sample

Table 7. Horse Power Generated and Outlet Temperature for various expansion efficiencies

Efficiency	Power	Outlet Temperature
%	btu/h	
100	593615.0	23.14
95	536294.0	28.33
90	481338.0	33.61
85	429352.0	38.91
80	380336.0	44.22

Figure 6 shows the phase diagram for the Dry Gas sample with wellhead and outlet conditions, and with varying expansion efficiencies. Note that at 100% efficiency, the gas is very close to the two-phase region.
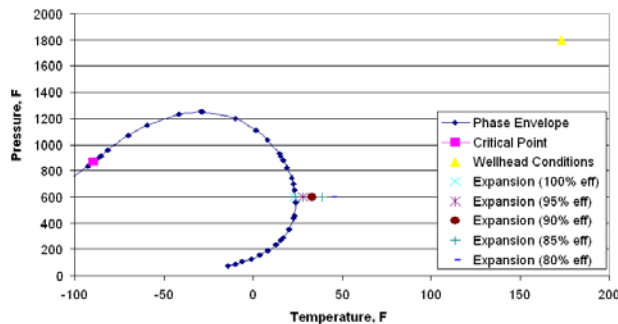

Figure 6. Phase Diagram for Sample 2

Therefore, the selected expander efficiency must allow the expansion of the gas to the required hydrate formation conditions and ensure that the gas remains in the single gas phase after expansion. If the gas sample does not remain in the single phase after expansion (two phase region), then this gas sample is not an appropriate candidate using this method. Additional separation facilities would then be required.

4. Hydrate Formation

The next stage after gas expansion/cooling is the hydrate formation. Natural gas from the expander, together with cold water, enters the reactor vessel at 35°F, and approximately 29,831ft^3 of hydrate is formed. The formation of natural gas hydrate yields a high latent heat of formation which must be removed to prevent dissociation. For 5 MMSCF of natural gas, 3.15×10^8 Btu must be removed. To achieve this, the formation vessel could be equipped with heat exchange tubes extending the full length of the vessel to enable effective removal of heat through the vessel. Figure 7 shows the end view of the vessel with heat transfer tubes.

Heat from the surrounding can transfer into the formation vessel and increase its temperature, causing dissociation of the hydrate as it forms. Insulation (e.g. NanoPore) is therefore necessary to minimise heat transfer with the surroundings. NanoPore thermal insulation provides exceptional performance with a very low overall thermal conductivity of 0.004 btu.in/ft^2.h.F. Because of its unique pore structure, NanoPore thermal insulation can provide thermal performance greater than conventional insulation materials (www.nanopore.com). About 240 btu/h of heat gained from the surroundings must be removed when using 1" thickness NanoPore material, compared to 4.35×10^7 btu/hr without any insulation.

It is proposed that the same vessel used to form the hydrate could also be used for storage and transport to its delivery point, whether the gas is for domestic use or international markets.
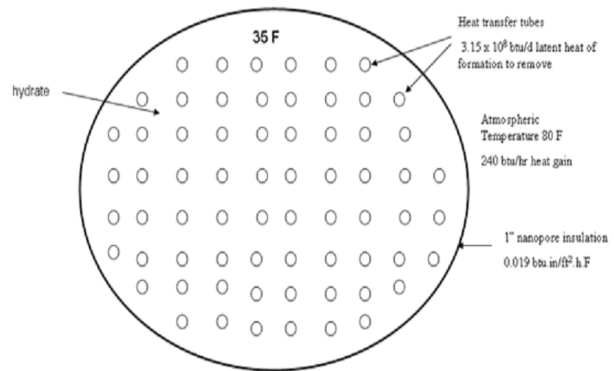

Figure 7. End View of Hydrate formation vessel with heat exchange tubes

The natural gas composition may have heavier components (C_5 or higher), as in the case of Sample 2. In the hydrate formation process, only C_1 to C_4 are captured, and the higher components (C_5 and higher) separate in the process as natural gas liquids (NGL). This is not the case with the Dry gas sample, which is primarily methane, with 1% ethane. Figure 8 shows the density difference between water, hydrate and NGL during the process.
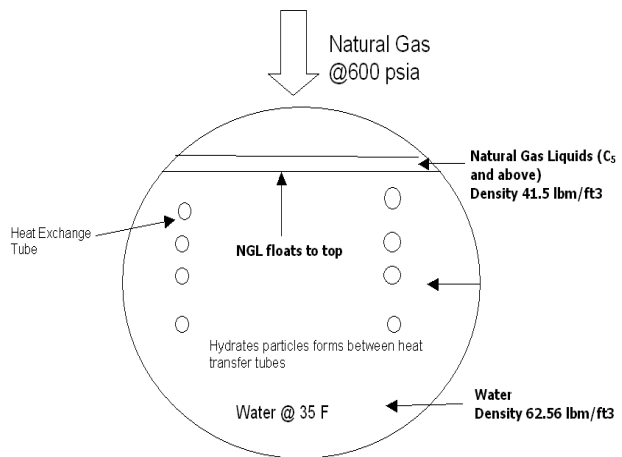

Figure 8. Density of Water, Hydrate and NGL

This separation method can be very useful for capturing lighter components necessary for efficient power generation. Some small amounts of higher components might have a negative effect on gas turbine systems. According to Ginter et al. (2001), there are problems with the presence of higher molecular weight components found in natural gas, as they can condense at low temperature, and appear as droplets in fuel supply, thus increasing the tendency for self-ignition. This also affects the flame position and combustion

stability. This is an important industrial application of the hydrate technology where heavy components from C_5 up are separated in the hydrate formation process. There is no need for gas processing prior to hydrate formation unless there is a significant amount of CO_2 or H_2S in the sample.

Once the hydrate formation is complete, temporary storage or transportation may be required.

5. Storage and Transportation

If gas storage is required, the same formation vessel could be used to store the hydrate. This is another potential application of gas hydrate technology where gas can be temporarily stored until later use. Depending on the storage time, additional cooling will be required to remove heat gain from the surroundings. Heat gain at a rate of 240 Btu/ hr must be removed. To this aim, the fixed heat transfer tubes in the vessel can be used, in combination with a small refrigeration unit. Figure 9 shows the conceptual storage system of gas hydrate, which can be used for possible gas storage for land-based power plants. Several storage vessels could be stacked side by side while awaiting hydrate dissociation to provide natural gas for uses such as power generation.

Transporting gas hydrate can be done both by land and by sea in order to deliver natural gas. This option uses the hydrate technology to transport gas to markets. A small refrigeration system may be required to remove heat gained from the surroundings. The same formation vessel can be used to transport hydrate using a trucking system (see Figure 9). For remote fields, where only railroad transportation is available, gas hydrate could be transported by train in the formation vessels.

For transportation by sea, the hydrate storage vessels could be placed in a container for protection, and transported to small-scale markets for short to medium distances (see Figure 9). According to Gudmundsson et al. (1998), natural gas hydrate transportation by sea is best suited for distances up to 12,000 km.
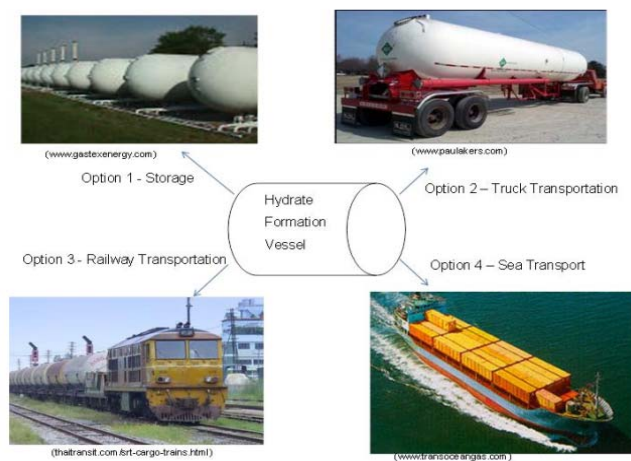

Figure 9. Conceptual Options for Storage and Transporting Gas Hydrate

6. Dissociation

The final stage in the proposed workflow is dissociation, when the hydrate conditions are altered to return natural gas and water. This dissociation stage is done using the heat exchange tubes to transfer heat to the hydrate, causing re-gasification.

The dissociation time at the market is another key aspect to the overall gas hydrate value chain. Dissociation of the hydrate can be done through depressurisation of hydrate or increasing temperature of the hydrate. In this study we only consider hydrate dissociation by increasing temperature. Faster dissociation rates would be facilitated by the heat transfer tubes that traverse the entire vessel. Therefore hot water can be pumped through the heat exchange tubes in the vessel (see Figure 7) to facilitate hydrate dissociation. The same amount of heat of formation removed during hydrate formation is required to dissociate the hydrate to gas and water. In this case, 3.15 x 10^8 Btu must be supplied to dissociate the 29,831 ft^3 of hydrate. The hot water is not transported with the hydrate but will be available at the market. There are several options using available seawater or disposed water. Water at 80°F can be appropriate to dissociate the hydrate (at 35°F) utilising the heat transfer tubes. In fact the analysis showed that the hydrate can be dissociated in 3.6 hours using water at 80°F pumped through the tubes. However, detail analysis of this will be published as another part of the entire study.

8. Conclusion

Several conclusions are drawn for the study. They are summarised below:

1) The expansion process yields useful energy that can be used in many ways, including power generation.
2) The expansion efficiency required to obtain the natural gas at 35°F may vary from sample to sample. In this case, for dry gas sample, 85% efficiency was required, whereas 90% efficiency was needed for sample 2. Some commercial expanders can have up to 90% expansion efficiency.
3) The expansion process must also ensure the gas sample remains in the single-phase region of the phase diagram, which is important in the design process.
4) Heat removal from hydrate formation can be achieved by using fixed tubes placed in the formation vessels. These tubes can also be used to remove heat gained from the surroundings during hydrate storage or transportation.
5) The fixed tubes can also be used to supply heat for subsequent hydrate dissociation.
6) The hydrate formation process separates out C_5 and higher as natural gas liquids, and captures C_1 to C_4. This is particularly important in cases where light

components are needed for power generation.

7) Gas hydrate can be used as a form of natural gas storage for future use, as in the case of power plants.

8) Insulation is required to reduce heat transfer into the vessel. NanoPore thermal insulation could provide superior performance for the vessel.

9) Using one vessel for hydrate formation, storage, transportation and dissociation could provide significant flexibility and hardware cost reduction.

10) Transportation of natural gas in the form hydrate by trucks, railway and sea can be considered especially in the absence of pipeline infrastructure.

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