

Technical Potential of Methods for Methane Extraction From Geopressured-Geothermal Fluids at High Temperature and Pressure

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Summary

Retaining pressure for direct-drive injection disposal of geopressured fluids is particularly cost-effective when combined with recovery of residual dissolved methane. We examine the technical feasibility of several chemical and mechanical techniques for extracting methane, and discuss specific chemical extractants, pros and cons of processes, and problems of implementation.

Introduction

Recent technical and economic assessments of methane (CH_4) production from geopressured-geothermal reservoirs in the Texas and Louisiana gulf coast area have been made by Swanson and Osoba¹ and Doscher *et al.*² They conclude that to obtain a reasonable return on capital for CH_4 recovery from reservoirs with properties shown in Table 1, the CH_4 selling price must be in the range of \$7.50 to \$9.24/Mcf.

Samuels³ also concludes that "unless the methane content and market value are sufficient to offset the cost of the production and reinjection wells, there is currently little incentive to develop this resource." It is clear that at the present value of methane (about \$3.50/Mcf), commercialization of the geopressured-geothermal resource will not be expected soon. Well capital costs comprise the major share of expenses, but the operating and maintenance (O&M) costs for injection are also significant. Doscher *et al.*² approximate the O&M costs for injection with the following formula.

$$\$/\text{bbl} = 2c + 0.005(\$/\text{Mcf})p, \dots\dots\dots(1)$$

where $\$/\text{Mcf}$ is the selling price of CH_4 and p is the required injection pressure in 10^3 psi. Pump power costs are proportional to injection pressure and the value of CH_4 . The $2c$ constant accounts for pump maintenance costs.

At 40 scf of CH_4/bbl brine, the value of CH_4/bbl brine is $0.04(\$/\text{Mcf})$. Assuming that pressure required

for injection into shallow aquifers (6,000 ft deep) is 1,000 psi, the ratio of injection O&M costs to the value of CH_4 is given by

$$R = 0.5/(\$/\text{Mcf}) + 0.125 \dots\dots\dots(2)$$

From Eq. 2, the injection O&M costs relative to the value of CH_4 are between 17.5% for \$10/Mcf gas and 26.8% for \$3.50/Mcf gas, clearly a significant percentage. The O&M costs are predominantly pump power and maintenance costs, which rise steeply with required injection pressure and become prohibitively high for geopressured waters with low CH_4 content.

It is clear that measures that could reduce injection costs would contribute significantly to the economic viability of geopressured resource development. One potential option in the production of geopressured aquifers is to maintain sufficient pressure at the wellhead to reduce the injection-pump workload. For shallow injection horizons, direct injection without pumping may be feasible. If it becomes necessary to inject into the production reservoir for pressure maintenance and subsidence control, injection-pumping O&M costs could easily exceed the value of recoverable CH_4 . Again, by efficient use of excess wellhead pressures, the power requirements for injection pumping could be reduced substantially. However, the potential savings would be offset by the loss of CH_4 still dissolved at the elevated pressures, as shown by the solubility curves in Fig. 1. Therefore, there is an incentive for extracting CH_4 at high pressures and also at production fluid temperatures to permit recovery of the thermal-energy component from the CH_4 -depleted brine.

There are several potential methods for extracting dissolved CH_4 at produced-brine temperatures of 302°F and at anticipated pressures of 1,000 to 1,500 psi for injection into 6,000-ft-deep aquifers. The chemical techniques include gas stripping, which is technically viable but may not be economical, and solvent extraction.⁴⁻⁷ Mechanical methods consist of positive-displacement

TABLE 1—PARAMETERS USED TO ARRIVE AT A PROFITABLE SELLING PRICE OF METHANE

	Study	
	Swanson and Osoba ¹	Doscher <i>et al.</i> ²
Reservoir pressure, psi	10,318	11,000
Drainage area, sq miles	12.6	11.0
Permeability, md	15	18
Porosity, %	20	21.6
Compressibility, psi ⁻¹	6×10^{-6}	1.1×10^{-5}
Thickness, ft	230	162
Well diameter, ft	0.458	0.400
Well depth, ft	15,560	13,000
Fluid viscosity, cp	0.20	0.236
CH ₄ /bbl fluid, scf/bbl	40	40
Average flow rate, B/D	9,700	9,956
Well life, years	20	20
Required profitable selling price	\$7.50/Mcf (includes recovery of 5¢/bbl for thermal-energy component)	\$9.24/Mcf (before federal taxes)

hydraulic pumps and hydraulic turbines coupled directly to pumps with provisions for CH₄ exsolution. Although the ensuing discussion addresses CH₄ extraction methods at pressures required for injection into shallow aquifers, these methods, in theory, should be applicable at the higher pressures necessary for injection into the production reservoir, and may be of greater value.

Gas Stripping

Gas stripping (or desorption) is a common chemical process for recovering dissolved gases in liquid streams. It should be possible to strip dissolved CH₄ from geopressured waters using this technique. To maintain pressure for direct injection purposes, desorption of CH₄ would be accomplished at injection pressures between 1,000 and 1,500 psi. The stripped CH₄ is recovered after separation from the working gas, while the spent liquid (still under pressure) is disposed of by injection without the necessity for pumping. The economic viability of this process depends on identifying a stripping gas that is either exceedingly inexpensive or has minimal solubility in geopressured brines.

Obviously, air would be the least-expensive gas-stripping agent. However, there are at least two critical limitations. First, the formation of an explosive air/methane gas mixture would not be acceptable. Second, the corrosiveness of the brine would increase greatly with aeration.

It should be emphasized that CH₄ solubility, even in pure water, is only 10 scf/bbl at 302°F and 100 atm.^{8,9} In 16-wt% sodium chloride (NaCl), CH₄ solubility at the same temperature and pressure is reduced significantly to about 5 scf/bbl.⁹ The lower CH₄ concentration would raise significantly the necessary profitable CH₄ selling price that was discussed above. At 10 scf/bbl, CH₄ is worth 3½¢/bbl (at \$3.50/Mcf). Therefore, the value of any stripping agent that is dissolved and presumably lost in the spent brine must be substantially less than 3½¢/bbl brine.

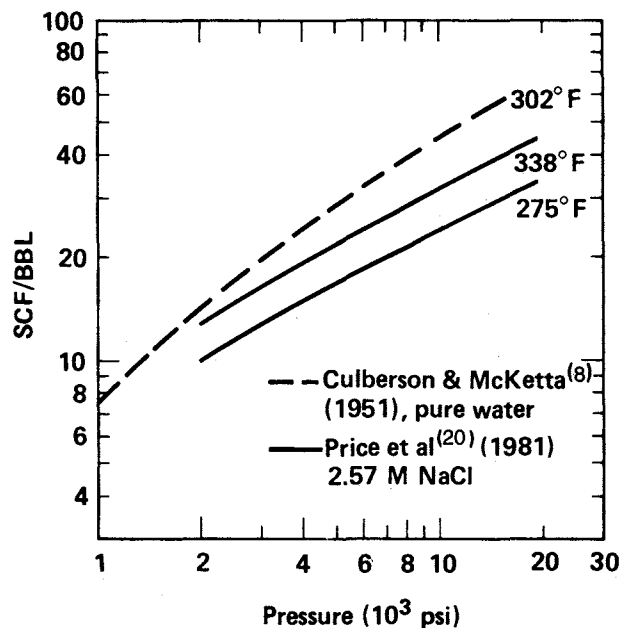


Fig. 1—Methane solubility in water and aqueous NaCl.

Another way to view the economics is to compare the value of CH₄ with that of bulk liquid nitrogen (N₂). The value of CH₄ at \$3.50/Mcf translates to 7.9¢/lbm, while liquid N₂ delivered in quantities greater than 1 MMcf/month costs about 4¢/lbm.* In short, because CH₄ is relatively inexpensive and its equilibrium concentration in the brine is so low, the choice of a stripping agent is critical and severely limited. Another consideration is the cost of the subsequent gas-separation process that is necessary to recover pipeline-quality CH₄, as well as the stripping agent for recycling. We have considered separation methods ranging from selective CH₄ absorption to condensation of either CH₄ or the stripping agent.

Halogenated Hydrocarbons

The first stripping agents we considered were the halogenated hydrocarbons, namely Freon™** refrigerants. Freon would be vaporized completely on contact with the hot brine, producing excellent gas/liquid interfacial contact for stripping dissolved CH₄. Gaseous Freon is condensed and separated from CH₄ at a temperature where the partial pressure of Freon in the recovered CH₄ stream is acceptably low in terms of cost and CH₄ purity.

However, very few Freon compounds have the desired thermodynamic and thermochemical properties to suffice as potential stripping agents. In addition, the stripping agents must be sparingly soluble in the geopressurized brines at temperatures of 302°F and at pressures of about 100 atm. They must be thermally and chemically stable in the brine environment. Finally, their critical temperatures must be less than 302°F to ensure complete vaporization for stripping purposes, yet they must have

*Private communication, Pacific Oxygen Sales Co. Inc., Oakland, CA (Feb. 1980).

**Reference to a company or product name does not imply approval or recommendation of the product by the U. of California or the U.S. DOE to the exclusion of others that may be suitable.

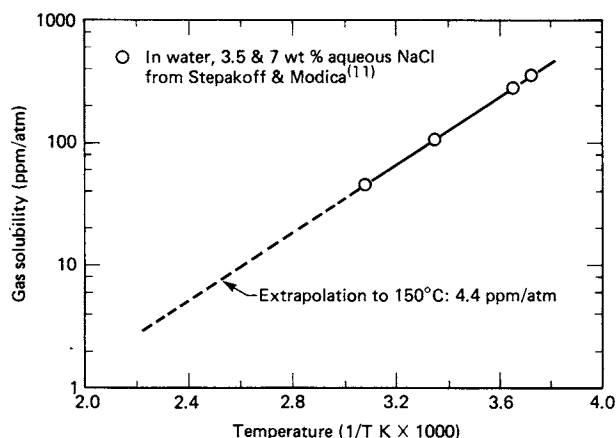


Fig. 2—Average Freon 114 gas solubility in water and NaCl solutions.

low volatility for ease of condensation in gas separation and purification.

The difficulty in selecting an appropriate stripping agent is partly because of the scarcity of solubility data in the literature at these temperatures and pressures. In our initial search, we found that dichlorotetrafluoroethane (Freon 114) met some of the criteria. Its critical temperature is 294.3°F and its normal boiling point is 38.8°F.¹⁰ However, at 100 atm and 302°F, its solubility in brine may be too high to be cost-effective. Stepakoff and Modica¹¹ measured the solubility of Freon 114 and other halocarbon refrigerants in aqueous systems containing as much as 7 wt% NaCl between 32 and 122°F. From the correlation given by Prausnitz,¹² we plotted the Freon 114 solubility data vs. the inverse of temperature on semilog coordinates in Fig. 2 and extrapolated to 302°F to give a solubility of about 4.4 ppm/atm. Assuming a Henry's law relationship is valid to 100 atm, the calculated solubility of Freon 114 at that pressure in 7 wt% NaCl at 302°F is about 440 ppm. At \$1.80/lbm for Freon 114,¹³ the value of Freon 114 dissolved in the brine and presumably not recoverable

exceeds the value of CH₄ eightfold.

Other halogenated hydrocarbons are not likely to be better. Partially halogenated hydrocarbons are considerably more soluble than completely halogenated hydrocarbons,^{10,11,14} so it appears that at a partial pressure of 100 atm only the cheapest gases, such as N₂, would even begin to approach economic feasibility.

However, relative process flow rates and number of stages in an ideal countercurrent operation can be determined for any stripping gas. An extrapolation of O'Sullivan and Smith's data⁹ on CH₄ solubility in 0- to 4-M NaCl at 124.7 to 257°F and 100 to 600 atm gives an average Henry's constant of 1.47×10^5 atm for CH₄ in 3-M NaCl at 302°F and 0 to 100 atm. Using this value to construct the equilibrium curve, we determined the number of ideal stages for a countercurrent multistage operation. With 0.56 lbm of Freon 114 per 100 lbm of brine, which corresponds to twice the minimum gas requirements, three or four ideal stages are required for 90% recovery of CH₄ from brine. The stage efficiency depends on the design characteristics of the stages and on the properties and flow rates of the fluids. For absorption or stripping of sparingly soluble gases, tray efficiencies can be as low as 10%.⁶ Therefore, a stripping operation may require towers with 30 to 40 stages. In practice, it may be more cost-effective to use fewer stages and higher gas/liquid ratios.

Similarly, it was determined that approximately four transfer units are required in a continuous differential contactor, assuming the worst case where the principal diffusional resistance resides in the liquid phase. The number of transfer units is an indicator of the ease or difficulty of the mass-transfer operation.

The gas-stripping operation with Freon 114 is illustrated in Fig. 3. The process flows merely illustrate relative quantities based on the equilibria data, 90% CH₄ recovery, and twice the minimum gas/liquid ratios that were used above for stage determination. Freon is condensed at a conservatively low temperature to minimize vapor losses in the CH₄ recovery stream and then is recycled.

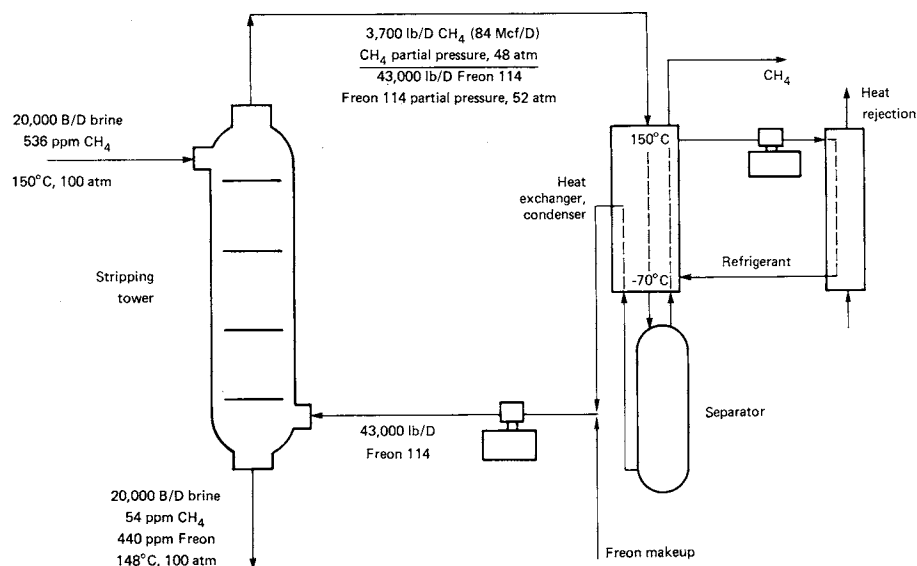


Fig. 3—Methane stripping from geopressured brines with Freon 114.

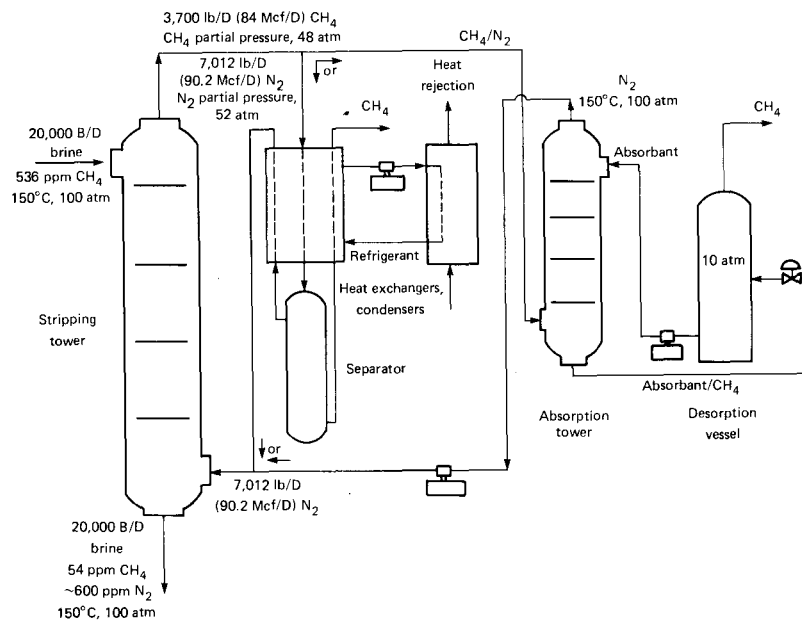


Fig. 4—Methane stripping from geopressured brines with N_2 gas. Purification by liquefaction or absorption.

Nitrogen

Liquid N_2 delivered by truck in quantities on the order of 10 MMcf/month costs approximately 4¢/lbm plus transportation. Large users able to absorb the capital costs of a N_2 pipeline can reduce the costs to perhaps 2¢/lbm. Although the cost of liquid N_2 is less than CH_4 selling at \$3.50/Mcf (7.9¢/lbm), its solubility at 302°F, extrapolated from data between 124.7 and 257°F,⁹ is roughly equal to that of CH_4 on a weight basis. Therefore, at 4¢/lbm, 50% of the recoverable CH_4 value would be lost by dissolution of N_2 gas in the brine. With these inherent losses, stripping CH_4 with N_2 is not promising unless CH_4 prices rise dramatically faster than liquid N_2 prices. This is a conceivable, but unlikely, event.

In the event that N_2 could be used, the stripped CH_4 could be separated from N_2 by preferential absorption with an appropriate hydrocarbon. Methane in essentially pure form would be recovered by simple depressurization of the hydrocarbon. Both the solubility of N_2 in the hydrocarbon and the vapor pressure of the hydrocarbon must be sufficiently low to minimize loss of N_2 and the hydrocarbon during the CH_4 recovery step. Low-vapor-pressure paraffinic hydrocarbons would be excellent absorbents for CH_4 . However, the solubility of N_2 in, for example, hexadecane, is also quite high¹⁵ and may be indicative of the lack of absorbents that are selective for CH_4 in mixed gases.

Fig. 4 illustrates the gas stripping operation using N_2 gas. The process flow rates are based on the same assumptions as the Freon 114 operation. As indicated, CH_4 could be separated from N_2 by liquefaction or by preferential absorption. However, liquefaction may be inefficient and energy-intensive because of the large fraction of noncondensable N_2 gas. Absorption of CH_4 would be feasible, provided an absorbent selective for CH_4 could be identified.

Solvent Extraction

The use of a paraffinic hydrocarbon for direct contact with the geopressured brines to extract dissolved CH_4 looks promising. The process is quite simple conceptually. A high-boiling, paraffinic hydrocarbon is contacted with the brine in an extraction tower. Methane, being more soluble in the hydrocarbon, is extracted and subsequently recovered in essentially pure form by depressurization of the extract. Solvent loss is controlled by selecting a low-vapor-pressure compound.

A promising hydrocarbon candidate is hexadecane, which has a vapor pressure of 10 mm Hg at 301.6°F and a solubility in water that is presumably quite low. We estimated hexadecane solubility using McAuliffe¹⁶ 77°F solubility data on C_1 to C_8 normal paraffinic hydrocarbons, and extended to C_{16} his interpretation that on an equal hydrocarbon vapor-pressure basis, approximately the same weight of paraffinic hydrocarbon dissolves in water. Therefore, the solubility of hexadecane was estimated roughly by multiplying the n-octane solubility at 77°F by the ratio of the vapor pressure of hexadecane at 302°F to the vapor pressure of n-octane at 77°F. This gave a value of about 0.5 ppm for hexadecane solubility at 302°F. Using this figure and a recent cost quotation of \$4/lbm for 95% pure hexadecane,* the value of hexadecane that is dissolved and nonrecoverable in the brine would be an acceptable 4.5% of the value of recovered CH_4 .

For the process to be feasible, the solubility of CH_4 in hexadecane must be significantly higher than in the brine to effect an extraction. Cukor and Prausnitz¹⁷ report Henry's constants for CH_4 in hexadecane in the temperature range of 77 to 392°F. Again assuming that Henry's constant is independent of pressure up to 100 atm, a solubility of 52,000 ppm is calculated for CH_4 in

*Private communication, Ventron Corp.—Alfa Div., Danvers, MA (Feb. 1980).

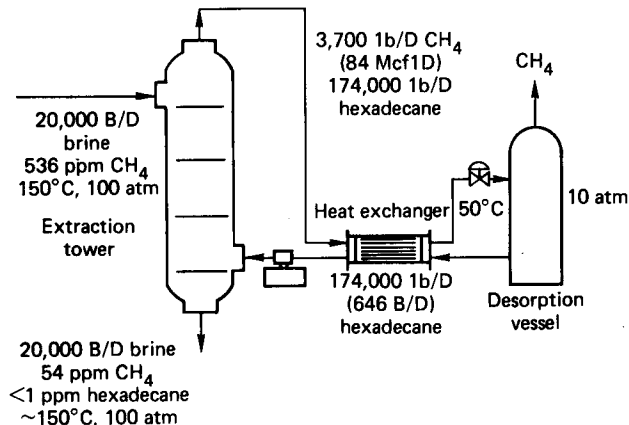


Fig. 5—Methane extraction from geopressed brines by liquid extraction.

hexadecane at 100 atm and 302°F. This compares with about 540 ppm for CH₄ in 16-wt% NaCl at the same temperature and pressure. Unlike gas stripping, solvent extraction can be more selective in removing CH₄. This would be significant in high-carbon dioxide (CO₂) brines, where removal of CO₂ could elevate the pH, initiating precipitation of dissolved minerals. But like N₂, CO₂ is also readily soluble in hexadecane.¹⁵

Again using the data of O'Sullivan and Smith⁹ for CH₄ solubility in 3-M brine, and those of Cukor and Prausnitz¹⁷ for CH₄ in hexadecane at 302°F, the number of ideal stages is determined for a countercurrent multistage operation for extracting CH₄ from brine. For 90% CH₄ recovery and 2.26 lbm of hexadecane/100 lbm of brine (corresponding to twice the minimum solvent requirements), three or four ideal stages would be required for the separation. As previously described for the gas stripping operations, stage efficiencies are highly variable. The number of actual stages will depend on stage design, fluid properties, and process variables.

For a continuous differential contact operation, we calculated that about three or four ideal transfer units would be needed. The number is weakly dependent on the phase (solvent or brine) that controls diffusional mass transfer.

Fig. 5 illustrates the liquid extraction operation for the conditions just described. The operation is quite simple in principle. However, the tower design can be quite complex because of potential problems that may arise from emulsion formation and, hence, difficulties in phase separation.

Formation of stable emulsion is a major concern in liquid/liquid extraction processes. Small bubbles of expensive solvent rejected with spent brine would be unacceptable. Low concentrations of surfactants may be necessary to assist in coalescing the dispersed phase. In addition, holding tanks may be required to allow settling, coalescing, and optimal phase separation. Upon

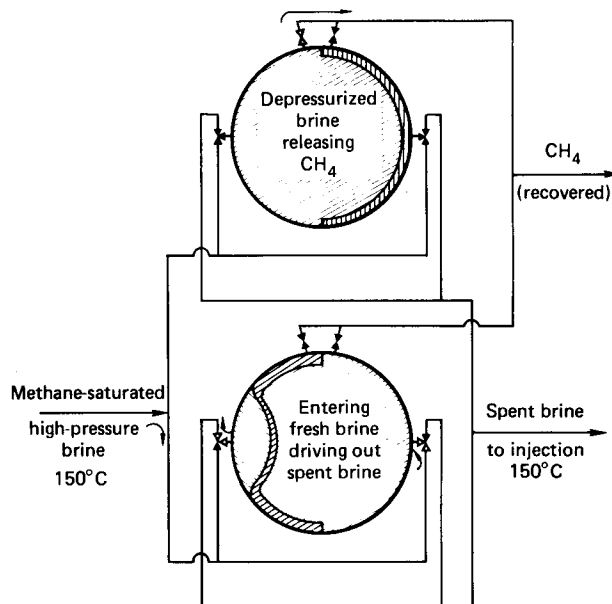


Fig. 6—Sequential depressurization/repressurization process for CH₄ desorption and brine injection in geopressed systems.

separation, CH₄ is recovered by simply releasing the pressure on the extract.

Subpilot Test Results

Recent measurements¹⁸ of 95%-pure n-hexadecane solubility in 15-wt% aqueous NaCl show that in the temperature range of 68 to 302°F, less than 1 ppm of hexadecane dissolves in the brine. Therefore, hexadecane would be a viable solvent from solubility considerations.

To demonstrate process viability, we recently conducted subpilot-scale liquid/liquid extraction tests using an Elgin-type spray column, 1½ in. in diameter and 6 ft high (results not yet published). Geopressed fluids were simulated with 15-wt% aqueous NaCl nearly saturated with dissolved CH₄ gas. The organic phase, n-hexadecane, was dispersed continuously as drops (about ½ in. in diameter) through the downward-flowing, continuous aqueous phase. Temperature and pressure test conditions were 302°F and 1,000 psi. Hexadecane/brine mass flow ratios ranged from 0.015 to 0.045. Preliminary analysis of the data shows that +90% recovery of CH₄ can be achieved at hexadecane/brine flow ratios of 0.040 to 0.045. Comparing the data with a stagewise countercurrent process, we estimate 1.5 ft as the height of an equilibrium stage for this column configuration. These initial test results indicate not only process viability but also high CH₄ recovery, with a relatively simple extraction-tower design.

Mechanical Methods for Methane Recovery

The simplest technique for recovering dissolved CH₄ is to lower the pressure to ambient and collect the evolved gases. The principal disadvantages, as previously mentioned, are the added pumping costs required for brine injection and the loss of thermal energy if pressure is fully reduced to 1 atm and, hence, about 212°F. Furthermore, if positive-temperature soluble species are pres-

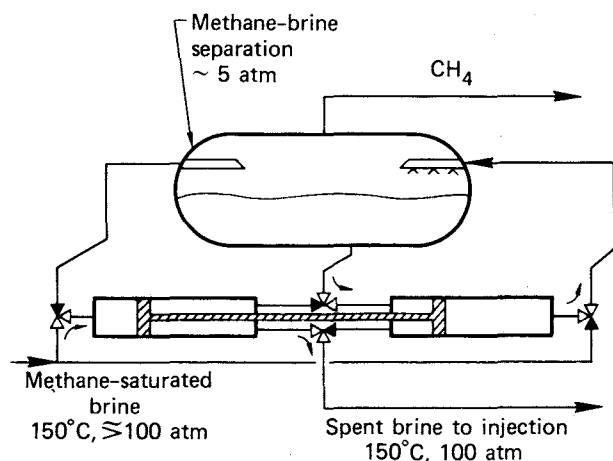


Fig. 7—Hydraulic-driven piston pump with a CH₄ separator.

ent, they may precipitate, creating fluid handling problems.

There are several potential methods that use the hydraulic energy available in the liquid discharged from the separator, which would operate at pressures somewhat below the wellhead but sufficient to drive the fluid-injection operation. The methods to be discussed are variations of positive-displacement hydraulic engines and hydraulic turbines, with special provisions to permit CH₄ exsolution and recovery.

Hydraulic-Driven Membrane Pump

The hydraulic-driven membrane pump is a sequential depressurization/repressurization process. A conventional gas separator is used for initial CH₄ separation from wellhead fluid. Pressure in that separator is adjusted to satisfy injection-pressure requirements, pressure losses in surface equipment, and pressure requirements that may be dictated by brine chemistry. Brine from the separator passes alternately through at least two depressurization/separation vessels in parallel. At any moment, one of the vessels is isolated from the flowing stream and the contained fluid is depressurized to no less than the vapor pressure of brine at 302°F (4.7 atm for pure water) to permit CH₄ exsolution without flashing of the brine. Concurrently, in the other vessel, brine (which has been depressurized already) is expelled to the injection well by displacement with fresh brine under pressure from the wellhead separator. To prevent mixing, a flexible membrane separates the spent and fresh fluid volumes.

When these steps are completed in the respective vessels, the vessel function is reversed to maintain continuous operation. Well flow is not interrupted, dissolved CH₄ is recovered, and direct injection of the spent brine without pumping is accomplished by proper valve sequencing. The process takes place at near isothermal conditions and does not preclude extraction of the thermal energy, if desired. Fig. 6 illustrates the basic flow streams and valving required for the sequential depressurization/repressurization process.

There are several aspects of the process that will require study and development. Methane exsolution rates

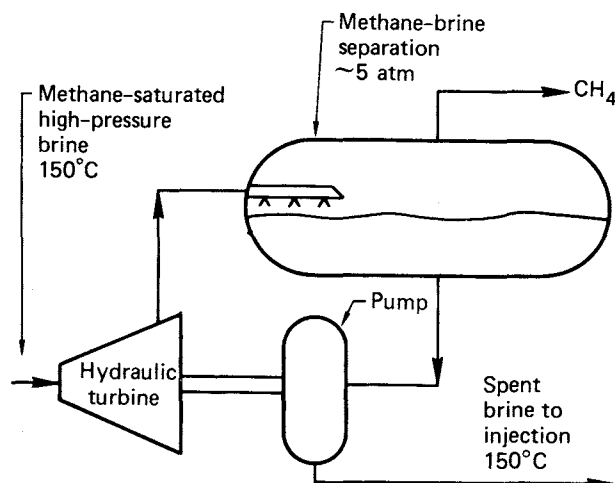


Fig. 8—Coupled hydraulic turbine-pump with a CH₄ separator.

and the dynamics of gas/liquid disengagement will require evaluation in laboratory tests. Mixing of the rich and spent brines must be held to an absolute minimum to ensure high overall CH₄ recovery. The valve-sequencing operation and the control system for depressurizing the vessels for CH₄ recovery must be developed. The most practical shape of the vessels may not be spherical but clam-shaped, which permits higher membrane cycling frequencies to reduce displacement volumes and, hence, vessel size. Whatever the shape of the pressure vessels, it is obviously desirable to minimize their individual size by using a gang of vessels. For example, if the total displacement volume cannot be reduced because of limitations in membrane cycling frequencies or the kinetics of CH₄ exsolution, then six pairs of 3-ft-diameter spheres cycling once per minute would pass 21,800 B/D.

Hydraulic-Driven Reciprocating Piston Pump

A coupled, dual-piston arrangement with a separate low-pressure vessel for CH₄ exsolution is shown in Fig. 7. This arrangement presumably overcomes the membrane-system difficulties. It separates the CH₄ exsolution step from the pumping operation and, by virtue of the piston-cylinder design, permits higher cycling frequency and lower pump displacement volumes. Again, ganged cylinders would be used to reduce cylinder displacement volume. A separate vessel for CH₄ desorption allows for more flexibility in handling gas exsolution problems.

Venturi Nozzle

The pressure regimes developed in a venturi nozzle may be used advantageously in this particular application. In a continuously flowing stream through the nozzle, the low-pressure regime at the throat would permit CH₄ exsolution, followed by liquid/pressure recovery on the order of 70% in a properly designed divergent section. Anticipated problems would be flow instabilities produced by continuous CH₄ exsolution in the converging section of the nozzle, gas separation and removal problems in the throat, and entrance flow instabilities in the divergent section. Scale formation in the nozzles also could be a serious problem.

Hydraulic Turbines

Other possibilities include use of hydraulic turbines or gear drives coupled directly to injection pumps, as shown in Fig. 8. Methane exsolution begins in the turbine chambers and is completed in a separator vessel. Velocity-pump reaction turbines and variations specifically modified for expansion of geopressured fluids are described by Austin and House¹⁹ and may be applicable in driving an injection operation.

All these devices have mechanical and hydraulic losses that need to be evaluated. The ultimate choice may not be based on the most efficient machine, but rather on the most reliable system capable of performing in the potentially corrosive and scale-forming geopressured-brine environment.

Conclusions

The costs required for conventional injection disposal by pumping can be eliminated or greatly reduced by retaining pressure for direct-drive injection. However, the cost benefits are severely eroded if that remaining portion of CH₄ still dissolved at injection pressures is not recovered. We have suggested and assessed several techniques for extracting CH₄ from geopressured fluids while retaining pressure.

1. Solvent extraction of CH₄ at pressure is a technically viable and promising process. Initial laboratory tests with n-hexadecane solvent and 15-wt% aqueous NaCl show a high recovery of CH₄ in a counterflow spray column using acceptable solvent/brine flow ratios.

2. Gas stripping is technically feasible, but does not appear economical because of gas dissolution losses and potentially high costs of separating CH₄ from the stripping agent.

3. Suggested mechanical techniques coupled with CH₄ separators are technically feasible, but much more in-depth analysis is required to ascertain engineering feasibility.

Acknowledgment

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SI Metric Conversion Factors

atm	× 9.806 650*	E-02 = MPa
bbl	× 1.589 873	E-01 = m ³
cp	× 1.0*	E-03 = Pa·s
cu ft	× 2.831 685	E-02 = m ³
ft	× 3.048*	E-01 = m
°F	(°F-32)/1.8	= °C
lbm	× 4.535 924	E-01 = kg
mm Hg	× 1.333 224	E-01 = kPa
psi	× 6.894 757	E+00 = kPa
scf	× 2.863 64	E-02 = std m ³
sq mile	× 2.589 988	E+00 = km ²

*Conversion factor is exact.

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