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Article Carbon-Negative Scenarios in High CO₂ Gas Condensate Reservoirs

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Abstract: A gas condensate reservoir in Northern Croatia was used as an example of a CO₂ injection site during natural gas production to test whether the entire process is carbon-negative. To confirm this hypothesis, all three elements of the CO₂ life cycle were included: (1) CO₂ emitted by combustion of the produced gas from the start of production from the respective field, (2) CO₂ that is separated at natural gas processing plant, i.e., the CO₂ that was present in the original reservoir gas composition, and (3) the injected CO₂ volumes. The selected reservoir is typical of gas-condensate reservoirs in Northern Croatia (and more generally in Drava Basin), as it contains about 50% CO₂ (mole). Reservoir simulations of history-matched model showed base case (production without injection) and several cases of CO₂ enhanced gas recovery, but with a focus on CO₂ storage rather than maximizing hydrocarbon gas production achieved by converting a production well to a CO₂ injection well. General findings are that even in gas reservoirs with such extreme initial CO₂ content, gas production with CO₂ injection can be carbon-negative. In almost all simulated CO₂ injection scenarios, the process is carbon-negative from the time of CO₂ injection, and in scenarios where CO₂ injection begins earlier, it is carbon-negative from the start of gas production, which opens up the possibility of cost-effective storage of CO₂ while producing natural gas with net negative CO₂ emissions.

Keywords: gas-condensate; enhanced gas recovery; CO₂ storage; CO₂ capture utilization and storage; reservoir simulation; carbon-negative

1. Introduction

The biggest anthropogenic CO₂ emissions contributors are fossil fuels combustion, iron, steel, cement, and fertilizers production and agriculture [1]. Being the biggest emitters, those industries are the very best candidates for carbon capture, utilization and storage (CCUS) technologies aiming at zero emission future. The first, and usually the most expensive step (60–85% of the overall costs) in any CCUS project is the CO₂ separation from the reservoir fluid stream [1]. The CO₂ sources are generally classified in two groups: (1) point sources (e.g., power plants, refineries, natural gas sources with CO₂ content ranging from 4 to 100% and (2) scattered sources (implies CO₂ capture from the atmosphere, where the concentration is significantly lower—400 ppm, or 0.04%).

Petroleum industry utilizes CO_2 for enhanced oil recovery (EOR), where CO_2 is commercially valued as raw material. However, due to high costs of CO_2 purchase, EOR incentives are adjusted to minimize the CO_2 use while maximizing the hydrocarbon production. If the incentive scheme changed, e.g., carbon charge, EOR economics would also change, and CO_2 storage estimation (potentially measured in Gt regionally) would become crucial, along with a simultaneous estimation of the commercial value of the produced hydrocarbons [2].

Carbon dioxide utilization in EOR processes has been successfully applied for over 40 years [3–5], and there are numerous reports regarding specific projects [6–9], and even specific problems [10,11], as opposed to CO_2 enhanced gas recovery (CO_2 -EGR), that



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). has been more intensively developed only in the last 20 or so years [12–14], especially for tight or gas-condensate reservoirs [15–17]. Some of the reasons for such a low level of the CO₂ injection research into natural gas reservoirs are high costs and high risk in production estimation. i.e., CO₂-breakthrough prediction. The injected CO₂ moves toward the injection wells and in this case, gas rate drops significantly, while the CO₂ production increases ([18,19]).

Physical CO₂ properties provide potentially feasible conditions for a reservoir repressurization [15,19–21]. CO₂ is 2–6 times denser in the reservoir conditions and consequently has lower mobility compared to methane, which should enable the CO₂ migration downward. This way, an effective methane displacement occurs. Solubility factor is another interesting property—CO₂ is potentially more soluble in brine than methane in reservoir conditions, and this feature delays the CO₂ breakthrough.

Natural gas reservoirs have significantly higher potential for CO_2 retention compared to depleted oil reservoirs, considering the same original fluid volumes in the reservoirs. Ultimate gas recovery reaches approximately 65% of the initial gas in place, which is almost double the usual primary ultimate oil recovery (35%).

Recently, attractive technologies have been those that ensure a secure gas supply in a carbon neutral, but more importantly, carbon-negative manner. It is usually necessary to analyze some complex concepts, such as Bioenergy with Carbon Capture and Storage (BECCS), or Direct Air Carbon Capture and Storage (DACCS). Such concepts encompass different CO_2 Capture and Utilization (CCU) or cogeneration technology sets, usually accompanied by a wide range of estimated necessary prices (penalties) of CO_2 emissions or emitted CO_2 quantities [22].

Since no similar example of analysis can be found in the literature, the motivation of this study is to show that the production process from a gas reservoir with extremely high CO_2 content (about 50%mol) can be carbon-negative when evaluated from cradle to grave. Except from the company's point of view, potentially carbon negative scenarios in already producing fields are attractive considering the technological and ecology frontiers as they encompass the use of highly efficient solutions that are in line with environment goals, i.e., CO_2 emissions reduction, which is currently not only a matter of isolated actions of few 'green' groups, but organized political effort recognized on an international level. An existing gas reservoir model was tuned (history matched) to real experimental and production data without increasing the complexity of the whole process, i.e., by converting a production well into a CO_2 injector. The results might help in the economic evaluation of the conditions under which the process could be cost-effective.

2. Materials and Methods

In the last 20 years, CO_2 -EGR related research has intensified, and within this work, a hypothetical-deductive research model [23] has been applied to test the hypothesis that carbon negative scenarios can be achieved in petroleum industry upstream. The quantitative methods used included the emission calculation according to published equations after different scenarios have been simulated to obtain the produced gas composition from a real geological model that had previously been validated. If only storage of CO_2 is considered, estimates of the CO_2 storage potential in depleted gas reservoirs can be considered simple, and estimates of CO_2 storage capacities can be reliably assessed by a simple (volumetric) material balance formula [24]:

$$V_{CO_{2(s.c.)}} = \frac{G_{i} \cdot B_{gi} - (G_{i} + G_{p}) \cdot B_{g_{CO_{2} + gas}}}{B_{g_{CO_{2} + gas}}}$$
(1)

where G_i is the initial (original) gas in place at standard conditions (m³, s.c. or sm³). G_p is the produced gas volume (sm³), $V_{CO_{2(s.c.)}}$ is the CO₂ storage capacity (sm³), B_{gi} is the initial gas formation volume factor:

$$B_{gi} = \frac{V_{g(r.c.)}}{V_{g(s.c.)}} \left[\frac{rm^3}{sm^3} \right]$$
(2)

where $V_{g(r.c.)}$ is volume of the gas at reservoir conditions and $V_{g(s.c.)}$ is the volume of same (number of moles) gas at standard (surface) conditions. $B_{g_{CO_2+gas}}$ is formation volume factor of CO₂ and natural gas (remained) in reservoir, at final injection conditions, and total molar composition of the system (mixture of original gas composition in the reservoir and CO₂) and can be calculated by some of commonly used cubic equations of state ([25–27]).

If data about produced gas are accurate, the above equations are useful for static estimates of CO_2 storage capacity as initial screening parameter. However, to estimate dynamic change of injectivity, more details about CO_2 injection strategy and about reservoir properties are required [28,29]. Injectivity mainly depends on reservoir permeability, heterogeneity and on reservoir and injection pressure. In zones where CO_2 saturation increases, relative permeability will also increase [30,31], which consequently increases injectivity, but the pressure will also be increased, which requires more energy for CO_2 compression and higher injection costs [32,33].

Immiscible process might occur when natural gas is displaced by supercritical CO_2 , which is described through effects of relative permeability [30,34], as more detailed (microscopic = pore scale or core scale) rock-fluid interactions, like miscibility, wettability and interfacial tension (of rock- CO_2 -natural gas-brine system) changes [35,36] are hard to include in (upscale to) reservoir-scale simulation models.

In this work, the Petrel RE (i.e., compositional Eclipse 300) reservoir simulation package was used (provided by Schlumberger), which makes it possible to include complex geological model and well models, and perform history matching with past production data (essentially well pressures and production rates, allowing validation of the spatial distribution of properties that are affecting multiphase porous flow).

Such dynamic models require data on relative reservoir geometry (including 3D fault modeling), porosity and permeability, and rock physics (relative permeability tables, capillary pressure tables, pore compressibility correlation, fluid compositions, and tuned equation-of-state parameters) and well data (completion data, skin factor, etc.). Since CO₂ is injected under high pressure, it is certainly in the supercritical state, which is treated as a liquid phase in the reservoir simulator. This allows some CO₂-natural gas relationships to be defined (the most important being relative permeability), and in this real case all parameters were used as tuning parameters for the history match).

By using such compositional simulator, detailed simulation of different future scenarios is possible, including (beside saturation and pressure change in 3D space with time) fluid compositions, among which gas composition is crucial as CO_2 emissions depend on the quality of the combusted gas considering its carbon content. The method of checking the carbon-negativity consists of calculating the overall emissions.

With known composition and volume of the 'pure' natural gas (gas from which CO₂ is removed) CO₂ emissions resulting from gas being used as fuel can be stoichiometrically calculated [37]: Produced gas (without CO₂) will be the source of new CO₂ emissions. Based on the known composition of the pure gas produced, these emissions can be calculated (assuming 100% efficient natural gas combustion) using the stoichiometric relationships [37]:

$$E_{CO_2} = V \cdot \frac{1}{\text{molar volume conversion}} \cdot C_c \cdot \frac{M_{CO_2}}{M_C} \cdot \sum_{i=1}^n M_i \cdot z_i$$
(3)

where:

 E_{CO_2} —amount of produced CO₂, kg V—(flaring) gas volume, m³

molar volume conversion—conversion from molar volume to mass (23.685 m³/kgmole) M_{CO_2} —molecular weight of carbon dioxide, g/mol

 z_i —molar fraction of component *i*, fraction

C_c—carbon content of the mixture:

$$C_{c} = \sum_{i=1}^{n} \left(w_{i} \cdot w C_{ci} \right)$$
(4)

w_i—weight fraction of component *i*,

wC_{ci}—carbon content of (hydrocarbon) component *i* (mass part of unit):

١

$$wC_{C_i} = \frac{M_C \cdot x}{M_i} \tag{5}$$

 M_C —molecular weight of carbon ($M_C = 12 \text{ g/mol}$)

x—stoichiometric coefficient for carbon (number of carbon atoms in a molecule) M_i _molecular weight of component, g/mol

)

For plus fraction, stoichiometric coefficient (x) is determined proportionally to its molar weight:

$$\kappa_{\rm C7+} = \frac{\rm M_{\rm C7+} - 2}{14.01} \tag{6}$$

The observed reservoir was modeled with a unique gas-water contact at an absolute depth of -2525 m, which was determined based on testing and analysis of well-logs (Figure 1).



Figure 1. Contour map of the formation top depths with the gas/water contact.

The model was spatially populated with porosity data (Figure 2) by the kriging method.



Figure 2. D porosity distribution model.

Before the development of scenarios for the purposes of this study, several preliminary checks were made on simplified models in terms of reducing the total number of cells and it was concluded that a satisfactory history match can be obtained with smaller models. For the needs of the dynamic model, a function is set to populate the model with permeability data and to include permeability anisotropy.

Function parameters were selected based on empirically proven relationships and performed and interpreted hydrodynamic measurements on wells of the reservoir, since laboratory core measurements are insufficient (too few data, i.e., 37 samples were measured from one well and 7 from the other, while other cores, if taken, were not used for petrophysical analyses). and they are not representative to be taken into account when determining reservoir permeability.

Reservoir fluid composition, which contains about 50% CO_2 , also contributes to the complexity of the system, which can be described as retrograde, gas-condensate. A sample from one of the wells taken before the start of reservoir exploitation was used to describe the fluid (Table 1).

Component	%mol
N2	1.98
CO ₂	48.95
C ₁	45.41
C ₂	1.66
C ₃	0.36
iC4	0.13
nC_4	0.22
iC ₅	0.07
nC ₅	0.07
C ₆	0.16
C ₇	0.15
C ₈	0.11
C ₉	0.10
C ₁₀₊	0.63
total	100.00

Table 1. Gas composition used in simulations.

Finally, history-matched model with small number of gridblocks (NX \times NY \times NZ = 28 \times 30 \times 9, which contains 7560 gridblocks—simulation cells) was used for simulating different scenarios of CO₂ injection.

The input data for the calculation of CO_2 emissions consists of a result data set for the base case (no injection at all) and result data sets for six simulation cases with simultaneous injection and production.

Production data for this gas reservoir are available for the period of 16.5 years, where the initial reservoir pressure (pi) of 396 bar at -2483 m (the reservoir brunt true vertical depth) has dropped to 51.5% of the respective pressure (simulation result, while the last measured data from the wells, and translated to average reservoir pressure at the brunt is 236 bar after 14.5 years). After tuning the reservoir parameters and obtaining a satisfactory quality of matching, the model can be used for different scenarios simulation and boundary conditions. The reservoir is rather small and developed with three production wells.

For overall CO₂ emissions analysis, two different EGR timings were chosen—early (EGR1) and late (EGR2). In order to compare the reservoir behavior with and without the application of enhanced recovery method, it was necessary to simulate the case in which the production continues in all 3 wells (after history matching of the first 8 years and validation of the model for the rest of the period for which real data exist). The base case was initiated at 0.75 of pi, which is the point in time for which real data still exist (and previous production and pressure are history matched). Adjusting the flowing bottom pressure in each well to 0.87 of the average (current) reservoir pressure (pr) resulted in

satisfactory matching of the cumulative gas production and pressure decline according to the next set of data. The rest of the production period (until the end of 40 years, which is a reasonable period for such a reservoir) is made of the base case scenario with the same drawdown. In the injection scenarios, two wells are kept as producers, while the third one becomes an injector. Injection is also defined through pressure multipliers (PM) for both EGR timings (Table 2).

Table 2. List of simulation scenarios.

EGR Scenario	Timing	PM (iBHP = $PM \cdot p_r$)
EGR1_30 EGR1_40 EGR1_50	0.75·p _i	1.3 1.4 1.5
EGR2_30 EGR2_40 EGR2_50	$0.5 \cdot p_i$	1.3 1.4 1.5

p_i—inital reservoir pressure; PM—pressure multiplier; iBHP—injection bottom hole pressure; p_r—current reservoir pressure.

3. Results and Discussion

Only one EGR scenario failed to reach the base case considering the cumulatively produced total gas (gas with CO₂), while 'pure' gas cumulatively produced showed suboptimal results of these development schemes (Figure 3). This can be ascribed to the reservoir fluid contamination, but the fact that there is one production well less should also be kept in mind.



Figure 3. Total and 'pure' gas cumulative production.

Regardless of the previously stated shortcoming of such scenarios, total CO_2 emissions were calculated for each of them.

Considering the high CO_2 content in the reservoir fluid, and assuming this CO_2 will be separated before gas is used (combusted), the original gas composition was normalized before emissions calculation (Table 3).

Component	y _i (%)
N2	3.88
$\overline{CO_2}$	0.00
C ₁	88.95
C ₂	3.25
C ₃	0.71
iC ₄	0.24
nC_4	0.42
iC ₅	0.14
nC ₅	0.14
C_6	0.32
C ₇	0.29
C_8	0.22
C9	0.19
C ₁₀₊	1.24
Σ	100.00

Table 3. Normalized composition of the gas to be used as fuel.

This composition eventually yields lower cumulative CO₂ emissions for all EGR scenarios comparing to the base case (Figure 4).



Figure 4. Comparison of CO₂ emissions in the base case and each EGR scenario.

For carbon 'negativity' assessment of the EGR process itself (cradle-to-gate), it is necessary to observe the difference between CO_2 cumulatively produced and injected (Figure 5). However, the most interesting result is the 'net' CO_2 obtained in each scenario, i.e., the CO_2 that would be emitted, or avoided by injection (Figure 6). This value is obviously dominated by the difference of produced and injected CO_2 trend, and negativity can be explained by, and ascribed to higher quantities of CO_2 injected than produced. Considering it was shown that in the case of high content in the original fluid, earlier EGR scenarios yield better results as significantly more CO_2 is injected than produced compared to the base case, they can be declared as carbon negative. Later EGR scenarios could be considered more adequate for reservoir conversion to a CO_2 storage facility as they result in higher 'pure' gas cumulative and lower total gas cumulative (Figure 3).



Figure 5. Difference between the produced and injected CO₂.



Figure 6. Total emitted CO₂ in each scenario.

 CO_2 enhanced gas recovery for the case of a typical natural gas-condensate reservoir in the Northern Croatia, with high CO_2 content (around 50%) mole was simulated to assess production possibilities in the transition period until zero-emission in 2050 are achieved in the European Union. This makes the analysis different from CO_2 -EGR studies—with a focus on CO_2 emissions, which is the opposite of usual EOR studies, where the objective is the maximum possible hydrocarbon recovery. Total amounts of CO_2 from both capture (which actually takes place in natural gas processing plants and thus represents one of the largest CO_2 point sources in Croatia) and combustion of the hydrocarbon portion of the gas produced were taken into account.

This is novel approach, as "pure gas" production was shown, and in this particular case, when more than a half of produced fluid (before CO_2 injection) is CO_2 such representation of results makes a big difference (Figure 3).

4. Conclusions

The production of pure gas in this particular reservoir can be optimized by shortperiod CO_2 injection, however this was not in the scope of this study. Features of the simulation results and the analysis are:

- all CO₂ injection scenarios result with lower total pure (hydrocarbon) gas production than base case (without CO₂ injection)
- lower cumulative hydrocarbon productions should be attributed to the fact that one production well is converted to CO₂ injection well, causing instantaneous production rate decrease.
- lower cumulative hydrocarbon productions can also be attributed to CO₂ breakthrough from injection to production wells and it can be connected to the fact that later start of CO₂ injection (EGR2 cases) results in higher cumulative production
- also, higher pure hydrocarbon cumulative production is observed for cases with lower CO₂ injection pressure (consequently, lower CO₂ injection rate)
- the increase in CO₂ production after breakthrough is smaller than CO₂ retention—the difference between injected and produced CO₂ is so large, that, from the beginning of CO₂ injection, all EGR case, except EGR2_30 become carbon negative (negative slope of EGR cases shown in the last figure)

The motivation of this study is to show that natural gas production can be carbonnegative if all emissions, even those from produced gas combustion are counted. Economic feasibility in this case will depend only on CO_2 compression costs and cost of conversion of one producing well (which is significantly less than for drilling a new one) as capital expenses and on fiscal system related to CO_2 capture, utilization and storage, as the production of natural gas at such field will be reduced. If stored and avoided CO_2 is counted as a value (negative) cost, CO_2 injection at natural gas fields becomes a great opportunity both to reduce CO_2 from the atmosphere and to produce natural gas costeffectively. Two main EGR strategies, one in early stage of production and one in later stage near depletion indicate also two opportunities for different business strategies

Early start of CO_2 injection would be feasible if CCS is to be acknowledged as the opportunity for storage of great amounts of CO_2 . In that case, the process should be recognized as utilization (carbon capture, utilization and storage, CCUS), as total CO_2 emissions from produced gas can be only a half of the CO_2 stored.

If CO₂ injection would start at near-depleted high CO₂-content gas reservoirs, that would make such gas fields carbon-neutral considering the cradle to grave cycle.

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