

Supercritical CO₂ injection in moderate-tight hydrocarbon reservoirs, a preliminary case study

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Summary

The research deals with the investigation of one alternative of carbon-dioxide utilization – underground storage – from a petroleum geoscience point of view. The basic assumptions and the results of the laboratory studies to be carried out later are based on a specific hydrocarbon field in Hungary. The previously measured and studied geological and petrophysical parameters of the reservoir (porosity, permeability, saturation, capacity, etc.) will be restudied and specified, based on the results of the new concept of laboratory experiments.

By defining these parameters, a 3D geological model, a “Dynamic model” will be created to understand the effect of carbon-dioxide injection on the dynamic behavior of a moderate-tight sandstone reservoir. Based on the results of the dynamic model, the storage capacity will be defined. The carbon-dioxide injection laboratory experiments will contribute to understanding these underground geo-chemical reactions (e.g. carbonation ability, compositional variation) and flow characteristics.

Keywords: carbon-dioxide injection, Carbon Capture and Storage (CCS), reservoir simulation, storage capacity, permeability

Szuperkritikus szén-dioxid besajtolása kis áteresztőképességű szénhidrogén tárolók esetén – előzetes esettanulmány

Rezervoár szimuláció CO₂ besajtolására és tárolására

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Összefoglalás

Korunk egyik legmeghatározóbb problémája az üvegházhatású gázok, különösen a szén-dioxid kibocsátásának csökkentése. Alapvetően két fő ágra oszlanak ezen törekvések: egyrészt a kibocsátásért felelős technológiák optimalizálása/„zöldítésére”, másrészt a már kibocsátott szén-dioxid hasznosítására (CCU), illetve befogására és letárolására (CCS). A kutatás a szén-dioxid hasznosítás egyik alternatívájával, a föld alatti letárolás lehetőségének vizsgálatával foglalkozik földtudományi aspektusból.

A kézirat alap felvetései és a későbbiekben magmintákon elvégzendő laborkísérletek eredményei egy konkrét, Magyarországon található szénhidrogén mezőre vonatkoznak. Emiatt más szén-dioxid tárolására alkalmas földtani képződmény (sókaverna, széntelep, illetve sósvizes aquifer) vizsgálatára a tanulmány nem tér ki. A laborkísérletek eredményeinek segítségével az előzetesen a mérnöki gyakorlatban használt és ismert tárolói paraméterek (porozitás, permeabilitás, kapacitás, telítettség stb.) kerülnek pontosításra. Az elvégzendő labormérések: higany besajtolásos

porozitás vizsgálat, centrifugális kapillaris nyomásgörbe és relatív permeabilitási görbék meghatározása, röntgendiffrakciós anyagvizsgálat. Ezen paraméterek ismeretében egy pontosított földtani modell kerül megalkotásra

A kutatás első fázisában a már meglévő, ipari gyakorlatban alkalmazott kőzetvizsgálati módszerek kerültek felülvizsgálatra, és egy új szemléletű, az eddigi módszereket pontosító eljárás került kidolgozásra a hazai geológiai formációkra vonatkoztatva. A későbbiekben a kőzetmintákon végzett tárolói paramétereket szimuláló szén-dioxid besajtolási kísérletek a föld alatti reakciók (pl. karbonátosodási képesség) és az áramlási sajátosságok megértéséhez és modellezéséhez is hozzájárulhatnak. Feltételezhetően a föld alatti reakcióknak köszönhetően egy, a korábbi becslésektől eltérő tárolótérfogatot lehet meghatározni.

Megállapításra került, hogy a szén-dioxid szuperkritikus állapotban besajtolva, rétegvízzel rendelkező zárt rétegekben egybefüggő „csóvaként” vándorol a porózus kőzeteken keresztül, amely a gravitációs szegregáció következtében vertikálisan kitágul a fedőkőzet alatt. Az oldalirányú szén-dioxid expanzió folyamatát a folyadékok csapdázódása korlátozza. Ezért a többfázisú áramlás és csapdázódás alapos vizsgálata elengedhetetlen a tárolókapacitás pontos meghatározásának érdekében.

A szén-dioxid besajtolhatóságát és a tárolási kapacitást nagymértékben befolyásolja a szén-dioxiddal telített sóoldat relatív permeabilitása, amely erősen függ a kőzet heterogenitásától. *Miljkovic (2006)* sóoldattal telített homokkő minták szimulációs méréseit hasonlította össze, amelyek csak a heterogenitás tekintetében tértek el egymástól. Megmutatta, hogy a kis strukturálatlan heterogenitás, úgy tűnik, nem befolyásolja jelentősen a CO₂ telítettségét és ennek következtében a tárolási kapacitást. Ezzel ellentétben *Kuo és társai (2011)* kimutatták, hogy a telítési profilt erősen befolyásolja a mag heterogenitása, és nagy injektálási sebességre van szükség ahhoz, hogy a heterogén közeg relatív állandó telítettségét elérje a homogénhez képest. Hozzájuk hasonlóan *Shi és társai (2009)* heterogén homokkő magokon szimulálták a szén-dioxiddal telített sóoldat elvezetését és beszívódását. Kimutatták, hogy a porozitás változása szignifikáns hatással volt a CO₂ migrációs mintázatára alacsony kiszorítási sebesség mellett és ez fokozatosan eltűnik az injektálási sebesség növelésével.

Fontos célkitűzésként jelenik meg a kutatásokban ezen tézisek vizsgálata, valamint a többfázisú áramlási kísérletek elvégzése a magyarországi CCS potenciális jelöltjeként számon tartott tároló magmintáin. A víz-gáz elvezetési relatív permeabilitási vizsgálatok szimulált tározó körülmények között szintén fontos új információkat fognak szolgáltatni, melyek lehetőséget teremtenek a CO₂ front végső eloszlásának meghatározására, valamint javaslattételre a tárolási kapacitás pontosítására és a geokémiai változásokra az adott tárolórétegekre jellemző heterogenitás függvényében.

Kulcsszavak: szén-dioxid-besajtolás, CCS, rezervoár szimuláció, tárolókapacitás, permeabilitás

Forewords

The scientific publication of Pál Gábor Veres KDP scholarship student reflects the main direction of his work. In Europe and globally, the greenhouse effect is becoming one of the most pressing problems of humanity, for which the carbon-dioxide emitted into the atmosphere can be blamed in a very significant part. To this day, researchers are divided on the question to what extent the currently experienced extreme climatic conditions can be linked to carbon-dioxide, but there is agreement that the amount entering the atmosphere must be reduced as soon as possible. The amount of energy obtained by burning fossil fuels and which cannot yet be replaced by alternative renewables at the moment cannot be removed from the life of mankind overnight. However, something must be done with the carbon-dioxide produced in the process. Carbon appearing in the research Capture and Storage technology can be one part of this process. It should be seen that the currently available geological data clearly show that only a small proportion of the carbon-dioxide generated can be stored in underground formations, but in the case of an industrial emission source, this can also represent a significant potential. The underground storage of carbon-dioxide is not a new thing, but the estimation of the

exact quantities that can be stored gives different results depending on the different methods. In the case of an actual industrial project, in which the payback time and emission quotas have a strong effect on the feasibility of the project, the question of the storable quantity becomes very decisive. A more accurate reservoir volume estimation process can greatly assist industry stakeholders in making a decision. The present research work adds to the most essential part of this determining process, the estimation of the potential, or may provide a precise method that was not available to specialists until now. Another important piece of information is that the validation of the method is performed on the basis of real samples of domestic, presumably carbon-dioxide storage, underground formations. Based on the multiphase flow experiments, simulations, and permeability tests to be performed during the work, it is expected to be possible to accurately determine the final distribution of the carbon-dioxide front, thereby specifying the storage capacity. I respectfully recommend the publication to the attention of all professionals interested in earth sciences.

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Pál Gábor Veres PhD student's paper represents one very significant element of a technology, the so called CCS (Carbon Capture and Storage) which is considered as inevitable both in EU and worldwide to reach the net zero carbon target by 2050. Recently more than 100 mega and big CCS projects are in the pipeline worldwide to reach more than 500 million ton/year CO₂ capturing till 2030.

The technology itself is known for more than 60 years, however, it is still in early stage. Despite the most critical, most expensive part, which is capturing CO₂ at the source, the last step of the technology, to store CO₂ in a deployed natural gas or oil field requires still substantial scientific work. This paper shows clearly how the simulation will be built up, which type of a physical, geophysical and other parameters need to be measured in a concrete, targeted storage field. The end result of simulation gives not only a good estimation of the size of storage facility but also the safety aspect of storing. There is a high need of public and regulators to have scientific guarantees of long-lasting, thousands of years safe operation, particularly storing.

Dr. Béla Kelemen

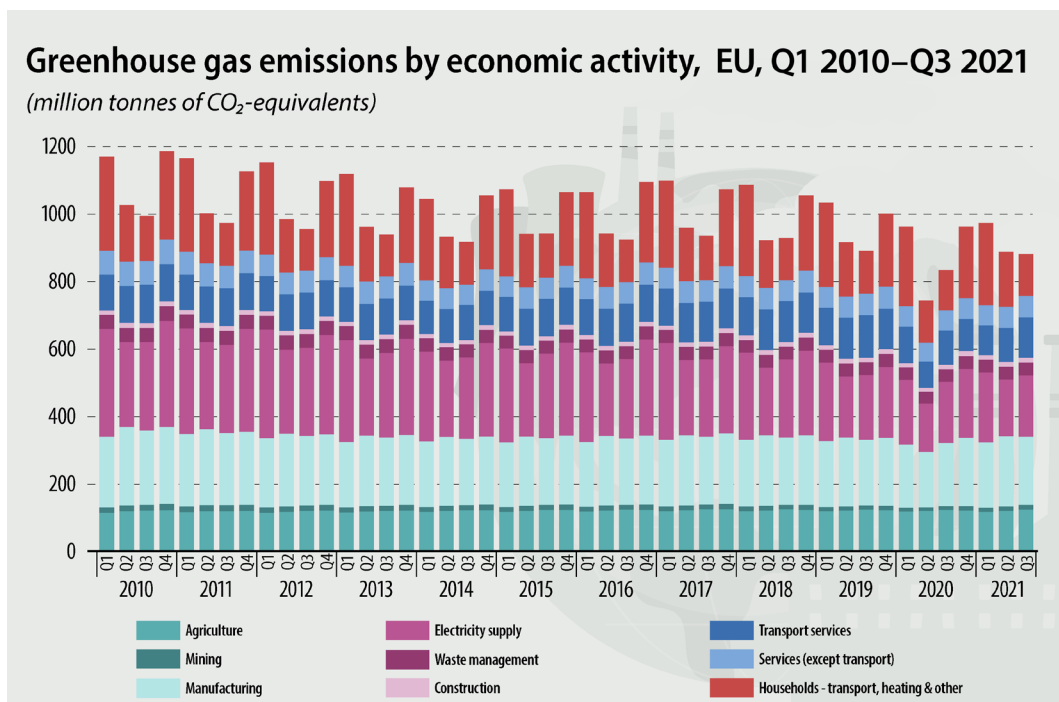
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The focus of public interest is most often the reduction of CO₂ emissions from human activities and transportation, residential and commercial buildings, but at the same time, the truly significant volumes – and hence the potential for a more significant impact – are linked to industrial processes (Figure 1). (Veres 2021)

Technologies for decreasing the CO₂ emission already exist, such as, without claiming to be exhaustive changing fossil fuels for renewable sources, boosting production and energy efficiency, implementing Carbon Capture and Storage (CCS) technologies, and discouraging carbon emissions by putting a price on them (see Figure 2). Although the commercial implementation of CCS technologies requires high capital and operating costs, advanced carbon capture technology, and carbon conversion and utilization can significantly reduce the cost of the carbon capture process and transform the captured CO₂ into a value-added product, which could improve the economics of the CCS system. The CCS is identified as an effective solution for mitigation of the CO₂ emission. The capturing involves carbon-dioxide at stationary point sources, which are single localized emitters, such as fossil fuel power plants, refineries, industrial manufacturing plants, and heavy industrial (iron, steel and cement) plants, as well as mobile sources, such as automobiles, ships, aircraft, etc., or directly from the air (Direct Air Capture). The captured CO₂ is compressed and transported for its storage in geological formations or for direct (non-conversion of CO₂, such as enhanced oil recovery, food and beverage, heat transfer fluids, etc.)

Introduction

The European Union, and a larger part of the world's developed countries have set quite ambitious goals for the coming decades in terms of decarbonization efforts.



ec.europa.eu/eurostat

Figure 1 | Greenhouse gas emissions by economic activity, EU, Q1 2010–Q3 2021

Source: Eurostat, 2022

EUA (EU ETS) Futures Prices



Figure 2 | The latest data on EU ETS carbon prices
 | Source: EU Carbon Price Tracker, 2022

and indirect (conversion of CO₂ into chemicals, fuels, and building materials) use.

The most challenging parts from technical and environmental points of view are capturing and storage. (Concawe 2020)

The CO₂ is injected into deep geological formations (e.g., depleting oil and gas reservoirs, deep saline aquifers, and unmineable coal beds) in supercritical condition and it is expected that CO₂ will be accumulated and trapped under the impermeable layer of caprock for a long period of time, hundreds to thousands of years. Basically, there are four CO₂ storage mechanisms (IPCC 2005):

(1) Structural and stratigraphic trapping: the injected CO₂ occupies the pore space and penetrates up through the porous rock and eventually accumulates beneath the very low permeability layers (caprocks).

(2) Residual trapping: when the CO₂ is injected into a formation, it displaces the formation liquid through the porous rock and continues to migrate as a separate phase because some of the CO₂ will be left behind in the pores and trapped there as residual CO₂.

(3) Solubility trapping: the injected CO₂ is dissolved in the formation liquid (e.g. brine) by the molecular diffusion such that it no longer exists as a separate phase (no buoyant forces which can drive it upwards).

(4) Mineral trapping: when the CO₂ dissolved in the formation liquid, it forms a weak acid that will enter into a chemical reaction with the minerals surrounding the rocks. It is believed that the chemical reaction between the dissolved CO₂ and the minerals will take place from over days (for carbonate minerals) up to thousands of years (for silicate minerals). (Negara et al. 2014)

This study deals with geological storage concept, investigation, and examines at least three types of geological formations in a specific hydrocarbon field in Hungary. This research summarizes the necessary laboratory experimental works and data to create a 3D geological model, “Static model” and “Dynamic model”, to understand the effect of carbon-dioxide injection on the dy-

namic behavior of the moderate-tight sandstone reservoir. (Shi et al. 2009). Based on the results of the dynamic model, the storage capacity will be defined. Also, the carbon-dioxide injection laboratory experiments will contribute to understanding and modeling of these underground geo-chemical reactions (e.g. carbonation ability) and flow characteristics.

Hydrocarbon reservoir – Case study

Field description

The field was discovered by the TZ-1 well in 1966. Two free gas and three oil reservoirs are known in the field. The Oil-Water-Contact (OWC) depth of the dissolved gas containing oil reservoir no. 2 is 2,100 m Below-Sea-Level (BSL). The oil is accumulated in the fractured top of the Variscan metamorphic basement rocks. The density of the paraffinic-intermediate oil is 914 kg/m³, the dissolved gas content is 35 m³/m³. The combustible part of the gas is 75.8%. The CH₄ 68.6%, CO₂ 18.2%, N₂ 6.0%, the C5+ content is 20 g/m³.

The reservoir rock of the oil reservoir no. 1 is Middle Miocene conglomerate and sandstone. The OWC depth is 2,020 m BSL. The density of the intermediate type oil is 906 kg/m³, the dissolved gas content is 35 m³/m³. The combustible part of the gas is 70%, CH₄ 64.0%, CO₂ 21.5%, N₂ 8.6%, C5+ content is 20 g/m³.

The depth of the multiple oil reservoir with gas cap no. 1 is at 1,940 m BSL, the reservoir rock is Middle Miocene limestone and coarse clastic rock, and the underlying Variscan metamorphic rocks in the pre-Cenozoic basement. The oil is paraffinic-intermediate, its density is 909 kg/m³, the dissolved gas content is 55 m³/m³, the combustible part is 68.8%. The CH₄ content is 63.2%, CO₂ 22.6%, N₂ 8.6%, the C5+ content is 44 g/m³. The combustible part of the cap gas is 68.7%, the CH₄ content is 63.3%, CO₂ 22.6%, N₂ 8.6%, C5+ content is 57 g/m³. The density of the condensate is 754 kg/m³, the amount is 50–55 g/m³.

The Gas-Water-Contact (GWC) depth of the lower of the two free gas reservoirs is 1,348 m BSL, the gas is situated in Lower Pannonian sandstone. The combustible part is 95.6%. The CH₄ content is 88.5%, CO₂ 0.8%, N₂ 3.6%, the C5+ content is 34 g/m³. The density of the condensate is 714 kg/m³, the amount is 58 g/m³. The upper free gas reservoir is located at a depth of 1,322 m BSL in Lower Pannonian shaly sandstone, the combustible part of the gas is 95.0%. The CH₄ 92.9%, CO₂ 0.5%, N₂ 4.5%, the C5+ content is 22 g/m³.

The condensate quantity is 20 g/m³. TZ North, an oil reservoir with dissolved gas, was discovered by the TZ-E-2 well in 1985 at a depth of 2,173 m BSL OWC in the Early Palaeozoic, fractured metamorphic basement with a low amount of free gas reservoir above it in the Middle Miocene conglomerate. The density of the paraffinic oil is 882 kg/m³, the dissolved gas content is 15.2

m³/m³. The combustible part of the gas is 96.3%, the calorific value is 41.0 MJ/m³. The CH₄ content is 79.9%, CO₂ 0.6%, N₂ 3.1%, C5+ content is 27.5 g/m³. The free gas reservoir GWC depth is 1,983.5 m BSL, the combustible part is 93.2%, CH₄ 86.4%, CO₂ 2.5%, N₂ 4.3%, the C5+ content is 40.3 g/m³.

A free gas reservoir was discovered by the TZ-North-6 well drilled near the well above at a depth of 2,048 m BSL (GWC) in 1985. The combustible part of the gas is 93.1%, CH₄ 83.9%, CO₂ 2.1%, N₂ 4.0%, the C5+ content is 122.3 g/m³. The TZ-E-9 well was drilled to the south-east of the two wells referred to above and uncovered a dissolved gas containing oil reservoir at a depth of 2,260 m BSL (OWC). The reservoir rock is Variscan granite, amphibolite. The density of the paraffinic-intermediate oil is 858.2 kg/m³, the dissolved gas content is

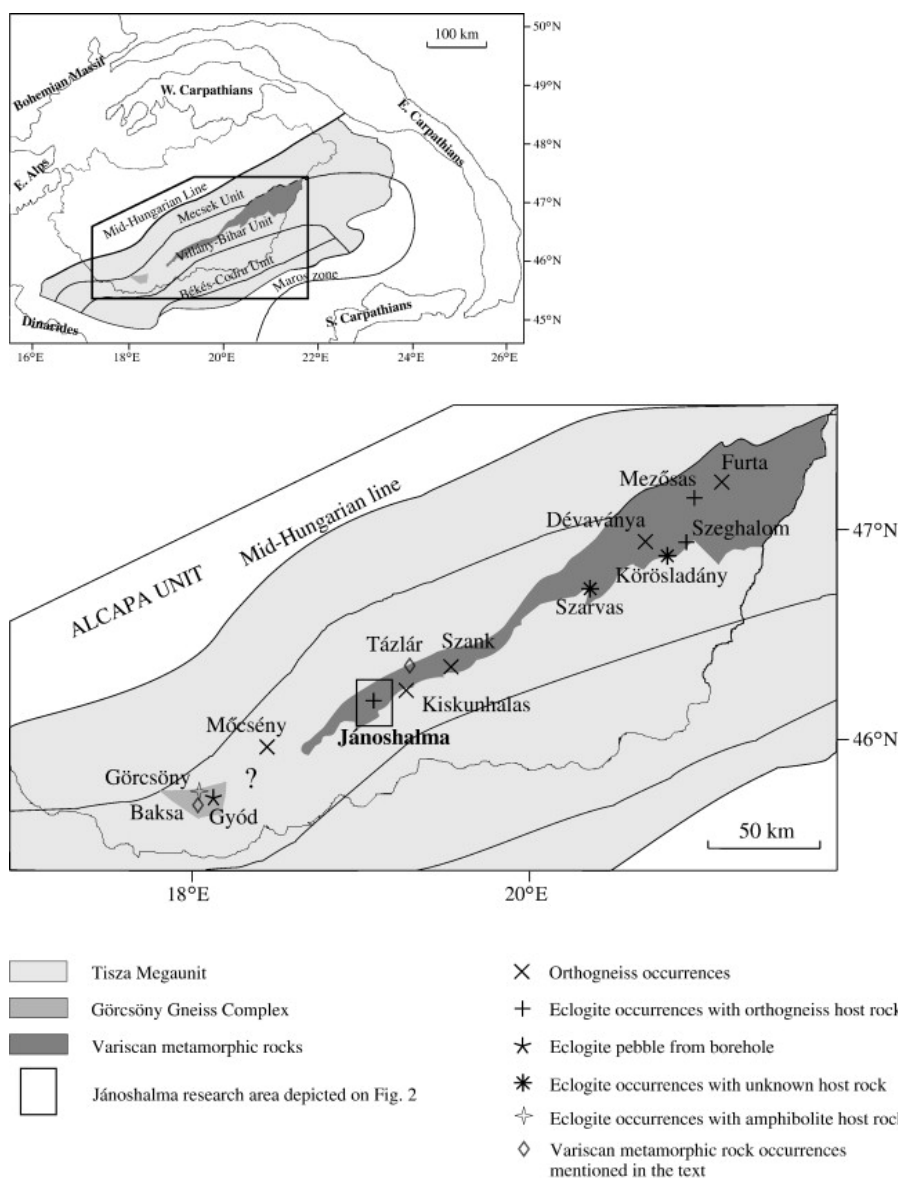


Figure 3 | Orientation of TZ field
 Source: Zachar et al., 2007

70.8 m³/m³, CH₄ 74.1%, CO₂ 6.2%, N₂ 2.2%, the C5+ content is 44.5 g/m³. (Kovács 2018)

The TZ Field (Middle Miocene conglomerate and sandstone) was selected to conduct the study on.

Figure 3 shows the location of this field in the Middle Miocene conglomerate and sandstone. The initial pressure was 217.7 bar and current pressure is 152.0 bar. The estimated oil production is 6.7% of the original oil in place (OOIP) under a moderated natural water driving force and gas solution and expansion with GOR 35.3 m³/m³. The actual Rock and Fluid samples have been collected and characterized.

Research methodology

This research will investigate the effect of several design parameters of 3D reservoir modeling of the field, including Special Core Analysis (SCAL) data, injection rate variation, hydrocarbon composition variation, storage volume capacity.

In order to conduct this study, the work will be divided into five phases:

Phase 1: Data collection and Preparation. It includes:

- Collect and prepare core samples.
- Collect fluid samples.
- Design and prepare the experimental setup (apparatus).

Phase 2: Determine the minimum miscibility pressure (MMP) for the injected gas.

Phase 3: Conduct flooding experiments storage CO₂ (CO₂ injection). Investigate the effect of the following parameters on injection rate, composition variation, storage volume capacity:

- A. Injection ratio.
- B. Size injection volume.

Phase 4: Conducting Reservoir-Modeling, Simulation work.

Phase 5: Results Analysis, Evaluation, and Interpretation.

Reservoir and wells selection

Core and fluid samples from the field have been collected. Porosity and permeability will be measured on all selected samples as a rock characterization main step at field level.

Core selection

The well logs restudied to locate the depth intervals for the rock samples selection are presented in Figure 4. The core samples selection procedure is shown in Figure 5.

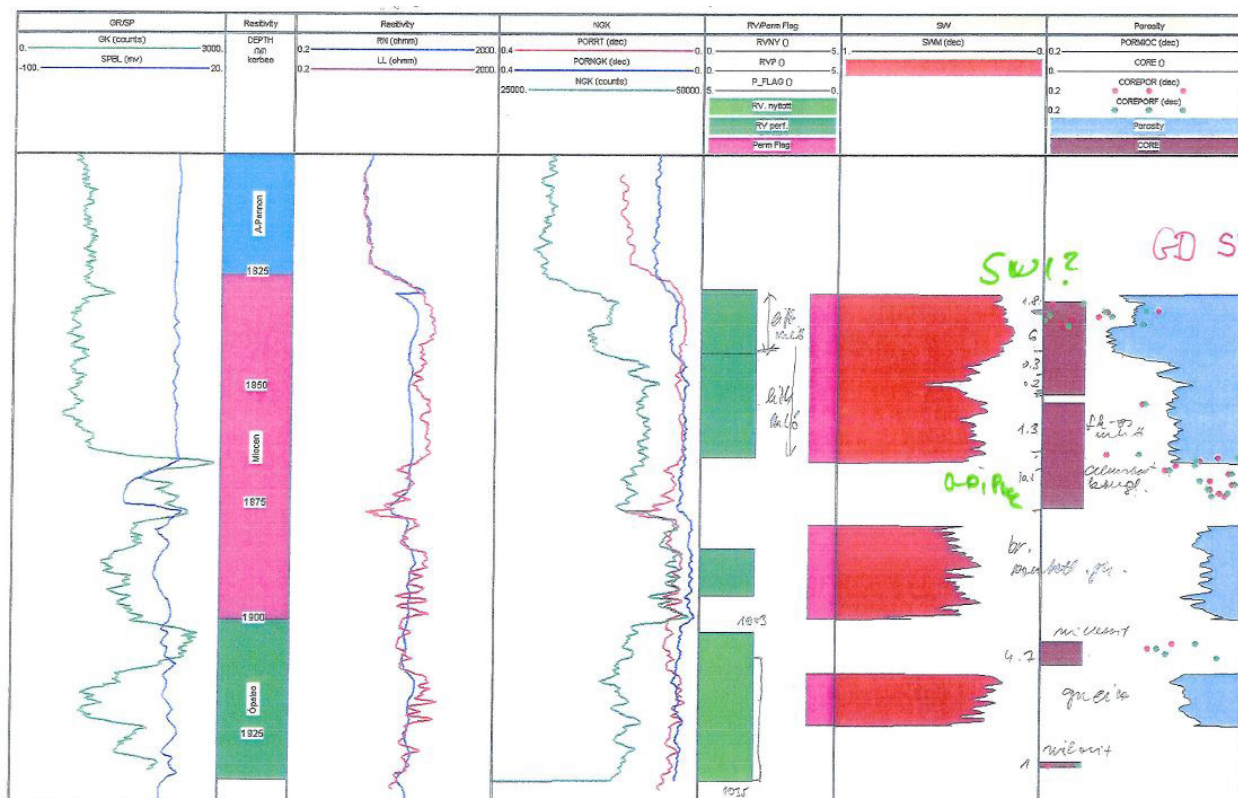


Figure 4 | TZ well log, selected intervals

Source: MOL Plc., 2022



Figure 5 | TZ field core selection procedure – Geology rock drill core samples in wooden box and prepared plugs
 | Source: MOL Plc. (author's own pictures), 2022

Core characterization

A diagram of the experimental setup that will be used in this study is shown in *Figure 6*. The core plugs will be cleaned and scanned dry by CT X-ray scanner to study the core plugs' pore structures. The porosity and permeability of the core plugs will be measured under reservoir conditions.

Figure 7 represents the injection-flooding unit with the device of X-RAY CT scanner that will be utilized to determine the porosity profile of selected core plug samples, also the relative permeability measurement, under

steady-unsteady state conditions, will be performed. The lab work will be designed and performed to handle a high pressure-temperature environment, reservoir condition.

In this research, the permeability will be measured under certain reservoir conditions, taking into consideration the pore pressure, overburden pressure and reservoir water salinity. According to the lack of continuous water sampling and monitoring from the field, the water salinity will be prepared artificially in order to simulate the formation water salinity. (Miljkovic 2006). In addition, for each of the core plugs to guarantee accurate

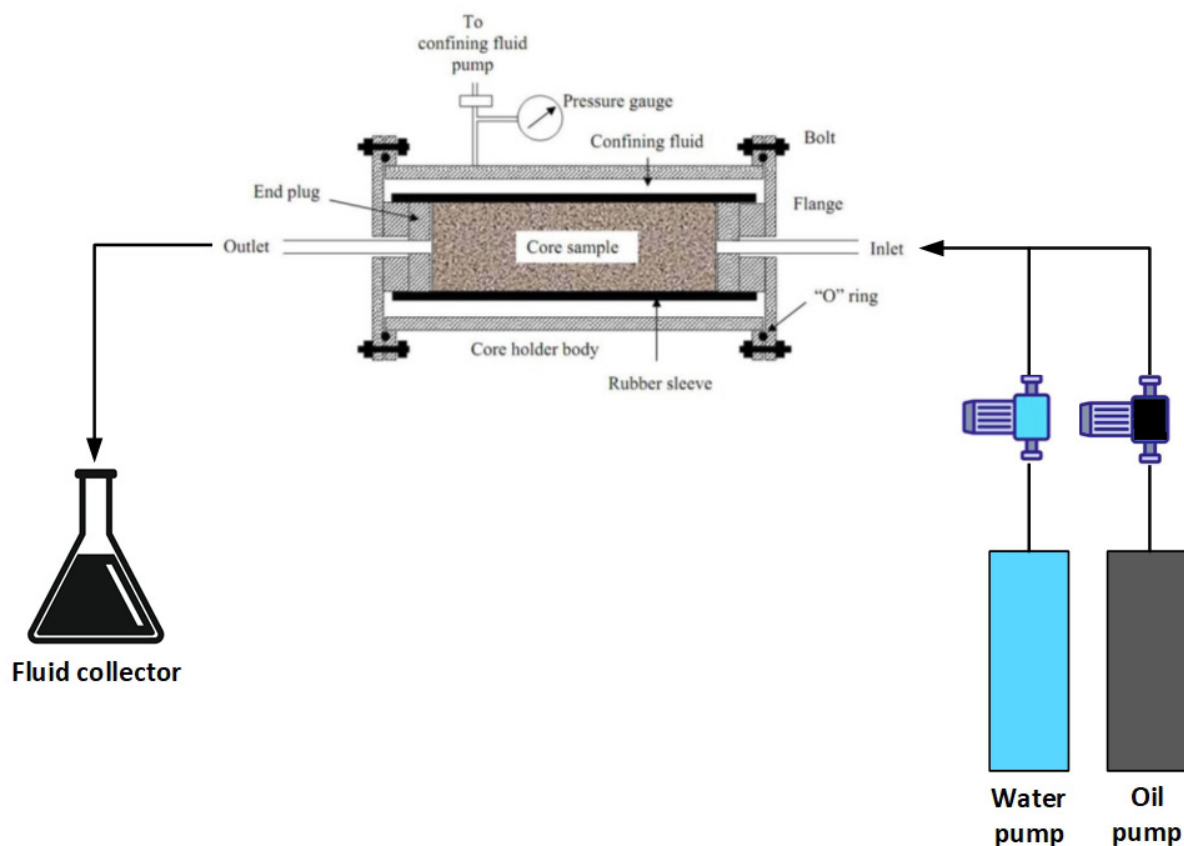


Figure 6 | Scheme of the lab experimental setup
 | Source: MOL Plc. (author's own picture), 2022

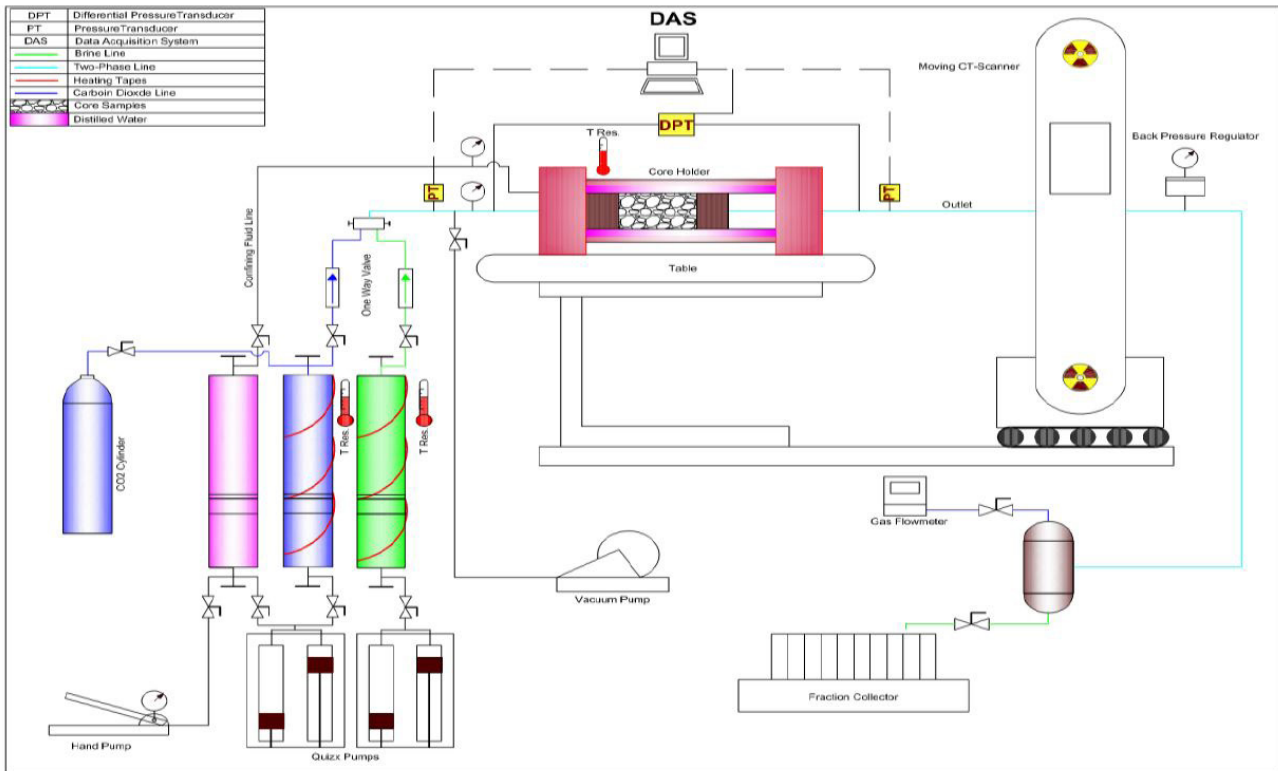


Figure 7 | CO₂ injection lab – Setup
 | Source: AlQuraishi et al., 2017

results, the permeability will be measured at different injection rates, then the average permeability value can be obtained for each of the core samples. (Kuo et al. 2011)

Each core sample will originate from different geological layers, the measured, summarized and evaluated properties should represent the whole reservoir specificity. The basic criterion of this phase is to make very exact measurements. After finishing the laboratory measurements, the derived results can be used to derive unified experimental based conclusions about the reservoir behaviors.

Numerical simulation

A numerical model will be created in this research for hydrocarbon reservoirs to evaluate the long-term storage of CO₂ in oil-gas reservoirs by taking into account typical compositions of dry, wet and condensate gases.

The reservoir simulations will be performed with an adaptive-implicit simulator Eclipse, or nonlinear regression analytical analysis, MBAL-Petroleum Experts software to investigate the macroscopic level mechanisms of the CO₂ injection and storage processes. The effects of several important parameters on the performance of injection and storage will be studied in order to optimize the CO₂ injection rate and storage volume capacity and to develop a set of screening criteria for selecting candi-

date reservoir layers for the development process. Based on the results of the simulation study, the conclusions will be drawn.

The numerical simulation parameters and the reservoir fluid properties, PVT are summarized in Table 1.

Table 1 | The reservoir simulation data that will be selected from several layers of the field

Porosity ϕ , [%]	Permeability – k, [md]
Connate water saturation – Swc, [%]	Oil Saturation – So, [%]
Gas Saturation – Sg, [%]	Reservoir Temperature, [°C]
Bed Dip Angle, [degrees]	Pay Thickness, [m]
API Gravity, [API]	Viscosity, [cp]
Bubble Point Pressure, [bar]	Gas-Oil-Ratio – GOR, [scf/stb]
Oil Formation Volume Factor at Bubble Point Pressure (Oil FVF at BPP), [m ³ /scf]	Water-drive Recovery, [%]
Original Oil in Place – OOIP [m ³]	CO ₂ Concentration, [%]
Radius – R[m]	
Temperature and pressure conditions resulted from natural state modelling	Existing wells design characteristics

Source: Dmour, 2010 (author's own edition)

Conclusions

The aim of this research is to investigate and examine the performance of CO₂ injection and storage in a Hungarian hydrocarbon reservoir. The simulation works will be carried out on the porous permeable sand/sandstone reservoir in the TZ area. A pure CO₂ will be injected into three formation types in order to simulate the reservoir dynamic behavior, composition variation effect, and to estimate the reservoir CO₂ capacity volume.

Based on the results of the experimental and simulation work, the following conclusions will be drawn:

- The reservoir may be assumed as multi-layered with continuous, horizontal layers, homogeneous by layers and with infinite extent.
- This simulation work will not take into account particle invasion and chemical transport phenomena.
- The main objective of the simulation is to investigate the hydraulic and dynamic behavior of the CO₂ injection well/reservoir interface (injection rates, volume capacities).
- To determine the best conservative permeability values for the reservoir, assumed varied injection reservoir thickness, is elementary to create a realistic model of the CO₂ storage/injection behavior.
- The primary/secondary porosity and mean permeability of the reservoir may considerably influence the injection pressure that will by all means increase during the processing time.
- The stimulation will be performed on a reservoir that will exhibit its ability to accommodate different CO₂ injection flows and injection pressures.

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