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## **CARBON DIOXIDE GEOSEQUESTRATION METHOD COUPLED WITH SHALE GAS RECOVERY**

### **1. INTRODUCTION**

Gradual warming of the earth's surface or the "globalwarming", resulting from the increased entrapment of solar radiation in the atmosphere by the 'greenhouse gases' is regarded as one of the most important environmental issues facing society. Although the estimates of this increase in temperature vary, depending on the models used for computation, an increasing average planetary temperature over the course of this century has been predicted by all these models. The temperature increase in the desert and frozen regions is believed to be even greater and may have a profound effect upon the vegetation and sea level in the near future [1].

The primary greenhouse gases causing global warming with their increasing concentration in the atmosphere are water vapor, carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), tropospheric ozone (O<sub>3</sub>). However, the highest relative contribution of these gases to the greenhouse effect comes from the anthropogenic emission of CO<sub>2</sub> (63.6%). It is estimated that the total emission of CO<sub>2</sub> is approximately 24 Gt per year [2].

Estimates of economic growth and associated emissions from the usage of fossil fuels as a provider of primary energy, suggest that the concentration of CO<sub>2</sub> in the atmosphere will continue to grow during this century unless significant steps are taken to reduce the release of CO<sub>2</sub> into the atmosphere. Four options are being explored to stabilize the atmospheric levels of greenhouse gases-increased conservation, use of less carbon-intensive fuels, adopting energy efficient methods, and carbon dioxide sequestration.

Carbon sequestration includes capturing CO<sub>2</sub> emitted from the combustion of fossil fuels and sequestering it, as well as reducing atmospheric concentrations by enhancing the

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uptake of CO<sub>2</sub> through natural sinks (e.g. forests, oceans, microorganisms). It is believed that carbon sequestration will enable the continued use of fossil fuels in energy systems without (or with reduced) emission of CO<sub>2</sub> [1].

Carbon dioxide capture and storage (CCS) is a set of technologies for the capture of CO<sub>2</sub>, its transport to a storage location, and its geosequestration (Fig. 1) [3].

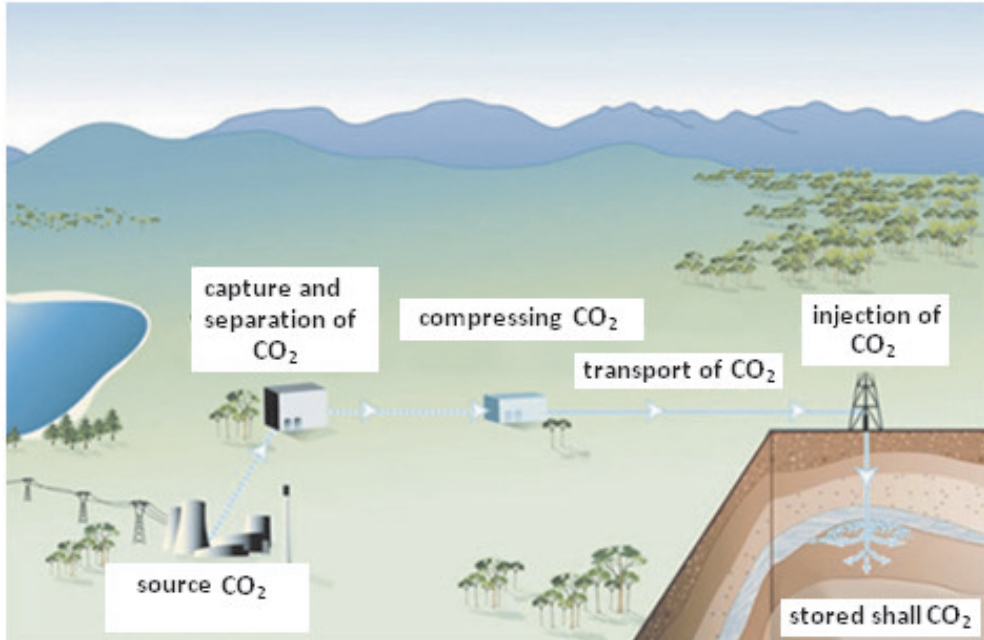


Fig. 1. Stages of geological sequestration CO<sub>2</sub> [3]

For the purpose of CO<sub>2</sub> sequestration can be used the natural environment, such as the Earth's ecosystem, the deep geological structures and oceans.

The main methods of CO<sub>2</sub> sequestration are as follows:

- terrestrial ecosystems imprisonment – sequestration in plants,
- sequestration in the ocean,
- mineral sequestration,
- geological sequestration [3].

Geological sequestration or underground storage of CO<sub>2</sub>, is one of the several possible solutions for carbon sequestration. Possible sites include deep unmineable coal seams, depleted oil and gas reservoirs, deep saline aquifers, abandoned and sealed mines, and so on. Among the possible geological sequestration options, storage within the deep unmineable coal seams offers one of the most attractive sites mainly on account of two reasons. First, coals exhibit a very high affinity for CO<sub>2</sub>, and second, the availability of huge coal resources

around the world especially in the same area as that of large fossil-fuel fired electric power generating stations.

Coals can store huge volumes of gases such as  $\text{CO}_2$  or  $\text{CH}_4$ , in a sorbed state, within the vast internal surfaces of micropores present within the coal matrix. Methane sorbed within the coal matrix, known as coalbed methane (CBM), serves as one of the important sources of clean energy throughout the world.

Geological sequestration assumes storage of  $\text{CO}_2$  in deep and permeable rocks, covered with impermeable tracks. Following underground storage space are considered: deep water levels, oil and gas deposits, deep and not exploited coalbeds.

Several conditions must be complied that can be geological storage of  $\text{CO}_2$  application (Herzog, Golomb 2004):

- the storage period should be long – preferably several hundred or more years,
- storage costs, including the cost of transport from the source to the storage area should be minimized,
- the risk of accidents should be eliminated,
- environmental impact should be minimal [3].

## **2. METHOD OF CARBON DIOXIDE UNDERGROUND STORAGE COUPLED WITH SHALE GAS RECOVERY**

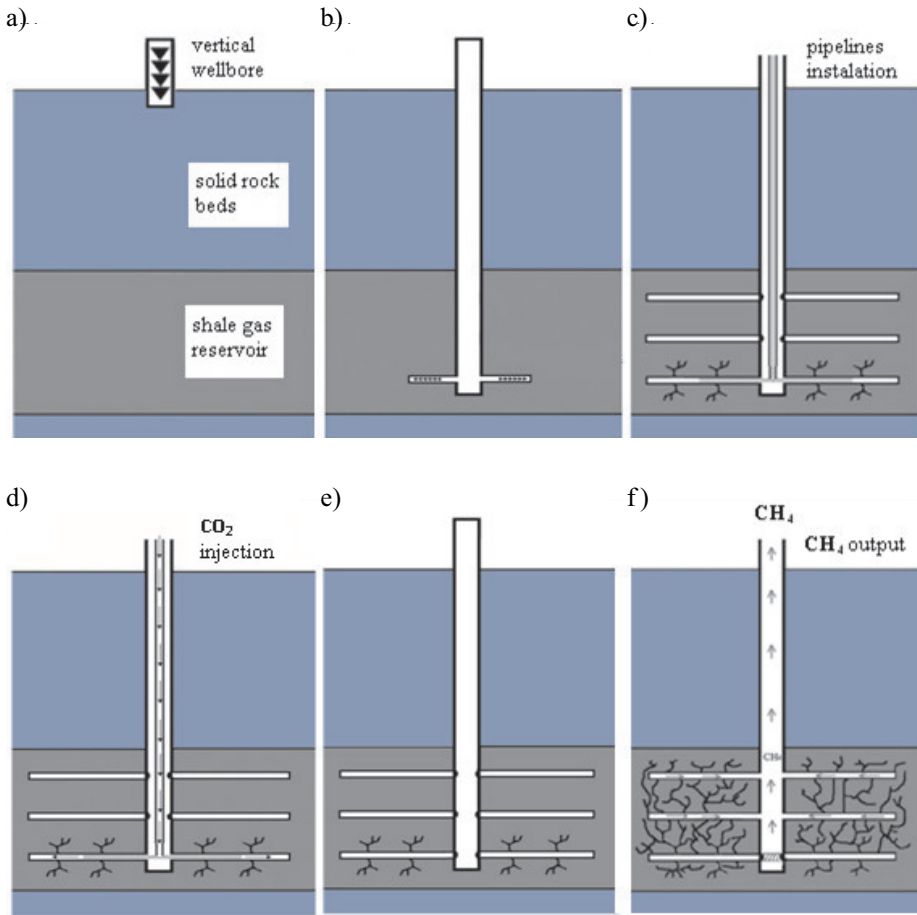
The presented method of carbon dioxide underground storage coupled with shale gas recovery (Fig. 2) was submitted by Military University as patent no P.398228.

The method of the shale gas recovery coupled with  $\text{CO}_2$  sequestration from the horizontal small-diameter wellbores made in single vertical wellbore was the subject of the proposed innovation.

The steps of the proposed method are presented below:

1. Firstly the horizontal wellbores have to be specially prepared in the shale gas deposit situated between solid rock beds (Fig. 2a). The existing horizontal wellbores can be also used.
2. Then the horizontal small-diameter wellbores are made circumferentially in a single vertical wellbore at a few depths (Fig. 2b).
3. The shale rock in the deepest wellbore can be initially perforated with the use of e.g. quasi-cumulative explosives. The upper not perforated horizontal wellbores are closed with the use of pins or valves. The elastic or half-elastic isolated or pre-cooled pipelines are installed in the open horizontal wellbores (Fig. 2c).
4. Then liquid cooled  $\text{CO}_2$  is injected to the shale gas reservoir with the use of cryogenic pump. During the injection the pipelines are progressively pull out from the horizontal wellbores for the precise filling of fractures. The  $\text{CO}_2$  injection process is finished after total pipeline pulling out (Fig. 2d). The  $\text{CO}_2$  injection process needs a continuous control of the temperature and pressure in the wellbore.

5. The open wellbores are closed with pins or valves controlled from surface. The thermodynamical process of heating cooled liquid  $\text{CO}_2$  in the reservoir is started. The process adsorption of  $\text{CO}_2$  and desorption of  $\text{CH}_4$  can last about 2 weeks (Fig. 2e). The temperature and pressure in the reservoir are controlled with the special set of sensors.
6. The upper not perforated horizontal wellbores are opened. The recovery of the shale gas can be carried out intrinsically (Fig. 2f). The whole process can be repeated for upper wellbores.



**Fig. 2.** Scheme of method of gas shale fracturing and gas recovery coupled with carbon dioxide storage

### 3. EXPERIMENTAL AND ANALYTICAL VERIFICATION OF THE METHOD

The method was verified on the base of analytical and experimental research.

The value of shale gas tensile strength can vary between 3 and 18 MPa (depending on the shale deposit location) under standard pressure. So if the pressure of CO<sub>2</sub> after heating in the deposit reaches that value, the method will be assumed to be correct.

The calculations were carried out with the use of REFPROP (Reference Properties) computer code developed by National Institute of Science and Technology (NIST). The code calculates the thermodynamic and transport properties of industry fluids and their mixtures with special consideration of cooling agents and hydrocarbons [4].

In the presented analysis, the equation of state used Span-Wagner.

The Span-Wagner equation of state was applied for CO<sub>2</sub> thermodynamic behavior description. The equation is an *empirical* representation of the fundamental equation of Helmholtz energy. Usually the dimensionless function of Helmholtz energy  $\phi = a/(RT)$  divided into an ideal gas part  $\phi^0$  and residual part  $\phi^r$  [5] is used:

$$\phi(\tau, \delta) = \phi^0(\tau, \delta) + \phi^r(\tau, \delta) \tag{1}$$

where:

$\tau$  – inverse of reduced temperature  $\tau = T_c/T$ ,

$\delta$  – reduced density  $\delta = \rho/\rho_c$ ,

$T_c$  and  $\rho_c$  – temperature and density at critical point.

The calculations performed for two states of liquid CO<sub>2</sub>: (-40°C, 2 MPa) and (20°C, 7 MPa). The analytical calculation results were presented in Figure 3. On the basis of those results it can be concluded that the final value of heated CO<sub>2</sub> exceeded the value of shale rock tensile strength (212 MPa for starting point -40°C, 2 MPa and 58 MPa for starting point 20°C, 7 MPa) and can cause its damage. In both cases was achieved the minimum pressure required for the cracking of the rock.

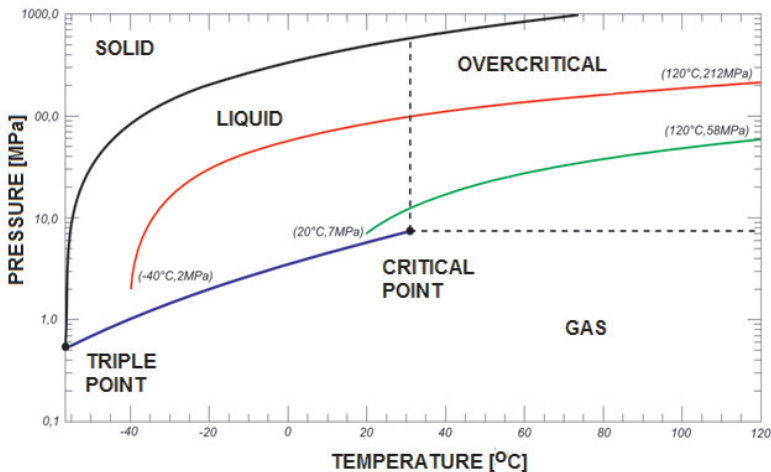


Fig. 3. Comparison of analytical and experimental tests of CO<sub>2</sub> heating

For the purpose of analytical tests verification the experimental test of isochoric CO<sub>2</sub> heating process was carried out. The CO<sub>2</sub> was used in the form of solid dry ice. It was heated in the closed pressure tank from the temperature of 23°C to 97°C. The temperature was measured with the use of cable sensor and pressure was measured with the use of extensometric tensor. The research equipment was presented in Figure 4.

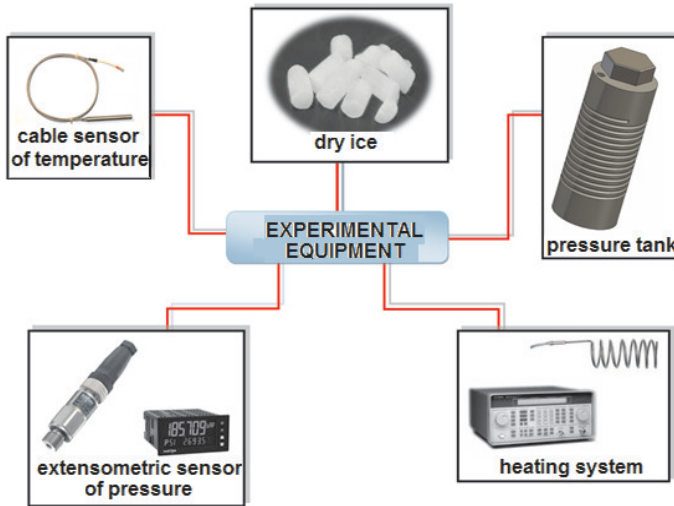


Fig. 4. Research equipment for CO<sub>2</sub> thermodynamic behavior testing

The comparison of both analytical and experimental tests was shown in Figure 5. The results showed good compatibility what proved the correctness of analytical method.

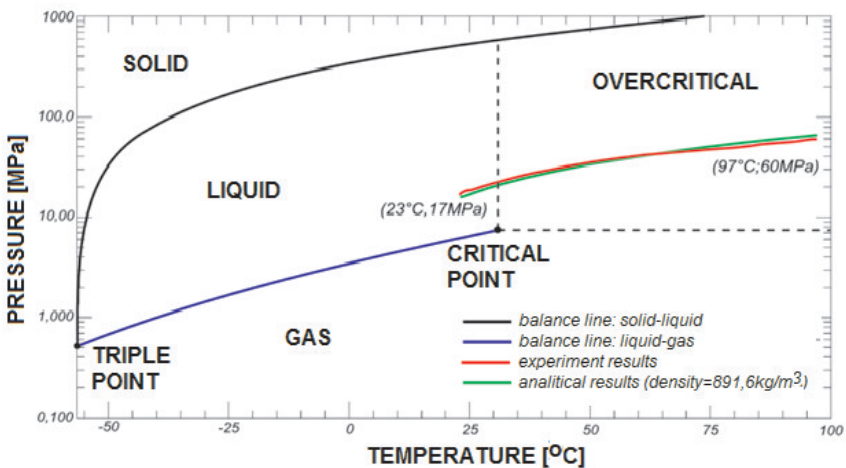


Fig. 5. Comparison of analytical and experimental tests of CO<sub>2</sub> heating

#### 4. CONCLUSIONS

1. The article describes an innovative method which allows for the efficient extraction of shale gas and carbon dioxide storage in shale rock.
2. The presented research results showed the possibility of liquid CO<sub>2</sub> use for shale fracturing and for damage of rocks at large depth.
3. The usage of liquid CO<sub>2</sub> can be an alternative for hydraulic fracturing replacing currently used water and chemicals.
4. The proposed method can be utilized for greenhouse gas storage after the shale gas deposit exploitation by closing the wellbore. It is an ecologically desirable effect.
5. Coal and shale gas deposits can adsorb two times more of CO<sub>2</sub> volume than CH<sub>4</sub> one. This property can be used for so called “clean energy” production (reached in closed cycle) – recovered CH<sub>4</sub> combustion for electric energy production near the wellbore and sequestration of CO<sub>2</sub> from that process.
6. There is a possibility of usage other greenhouse gases, that are heavier than CH<sub>4</sub>.
7. The method can be applied in various gas and porous rocks.

#### REFERENCES

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