Vol. 17, No. 4

ISSN 2064-7964

PERMEABILITY MEASUREMENT THEORY IN CASE OF NATURAL GAS AND NATURAL GAS-HYDROGEN MIXTURE

^{1,} *Ildi Bölkény, ²Marianna Vadászi, ³Ammar Saliby

^{1.3}Materials and Intelligent Technologies Higher Education and Industrial Cooperation Centre - EIKI, University of Miskolc, Miskolc, Hungary.

²Institute of Petroleum and Natural Gas, Faculty of Earth Science and Engineering, University of Miskolc, Miskolc, Hungary. e-mail: *bolkeny@eiki.hu , ²vadaszi.marianna@uni-miskolc.hu , ³a.saliby@hotmail.com

Received: 22nd February; Accepted: 30th August

ABSTRACT

Green hydrogen, using renewable electricity that breaks down water molecules into hydrogen and oxygen, holds great promise for meeting global energy demand while contributing to climate policy goals. Interest in green hydrogen production technologies has increased considerably. This is because the potential uses of hydrogen cover many sectors, including power generation, manufacturing processes in steel and cement production, fuel cells for electric vehicles, and power grid stabilization. One possible use of green hydrogen is to blend it with natural gas and deliver it to end-users using existing natural gas pipeline storage and networks, thereby increasing performance and reducing emissions. In the case of underground storage of a hydrogen natural gas mixture, it is important to assess its impact on the reservoir beforehand, which rock permeability studies can do. This article deals with the theory of rock permeability testing for natural gas and natural gas-hydrogen mixtures.

Keywords: natural gas, hydrogen, permeability

1. INTRODUCTION

The permeability of rocks is one of the basic parameters determining the dynamics of motion of natural and anthropogenic fluids in a geological medium [1,2]. Therefore, it is necessary to determine the permeability of rocks to simulate the processes of ore formation, develop underground hydrogen natural gas storage facilities, and solve many other basic and applied issues. Additionally, it is important to remember that the structure and arrangement of rocks, rather than their content, govern their permeability. These characteristics have sizable intrinsic variability. As a result, even rocks belonging to the same lithologic class can have permeabilities that differ by many orders of magnitude [3]. Rocks' permeability may be tested in a lab using samples, even at high temperatures and pressures consistent with their occurrence in deep layers of the Earth's crust. Research findings on the permeability of anisotropic rocks are particularly rare. From a practical perspective, the situation where the layered rock structure of a rock causes the anisotropy of permeability is most significant. The available experimental information on the permeability of such rocks is often derived from studies using cylindrical samples, where one axis is perpendicular to the plane of layering and the other lies in the layering plane [4,5]. The process and equipment for making very accurate measurements of the permeability of rock samples for a single-phase fluid and its anisotropy are examined. The Klinkenberg constant, a crucial aspect of the sample's pore space, and the value of permeability for a condensed fluid can both be extracted from the data of a single experiment using the transient method modified to account for the dependence of a percolating gas' properties on the parameters governing its state. The value of the axial and radial components of the sample's permeability may be calculated concurrently during a special experiment. Experiments can also be carried out at high temperatures and pressures.

DOI: https://doi.org/10.14232/analecta.2023.4.61-68

Vol. 17, No. 4

ISSN 2064-7964

2. TECHNIQUES FOR MEASURING THE PERMEABILITY OF ROCK SAMPLES

2.1. The steady technique

Typically, cylindrical samples are used for permeability studies in laboratories. Such a sample is tightly sealed along its side walls using the stationary flow technique. Its end is exposed to both constant and variable percolating medium pressure levels (gas or liquid) P_{in} and P_{out} , as shown in Fig.1, the permeability value is obtained using the Darcy law under the presumption that the vectors of percolation velocity across the entire sample are equal in magnitude and perpendicular to its end faces [6].

$$k = \frac{\mu. L. G}{S. (P_{in} - P_{out})} \tag{1}$$

Where μ is the dynamic viscosity of the percolating medium, L is the length of the sample, S is the cross-sectional area of the sample, and G is the flow rate.



Figure 1. The Steady technique [6]

Water is a slow-percolating fluid; therefore, measuring permeability takes much time. For percolation through a sample, gases were utilized to decrease the time of tests (fluids with a lower dynamic viscosity). By using gases (often argon or nitrogen), it was also feasible to prevent the study samples from changing unfavorably due to the water-rock interaction at high temperatures, particularly severe.

2.2. The oscillating flow technique

In this technique, the fluid pressure at the sample's input end face is a periodic function of time when permeability is assessed using the oscillating flow method, as illustrated in Fig. 2, The ratio of amplitudes and phase displacement of variations in the fluid pressure in the input and output reservoirs determines the values of permeability of samples [7,8]. According to its creators, the technique's benefit is the experiment's relatively brief runtime. This process may be modified to assess the sample's porosity and permeability. The percolating fluid might be either gas or water. High amplitudes in the pore pressure of a gas (approximately 1 MPa) are commonly utilized for these studies. Based on this, it has been claimed that the Klinkenberg effect has a negligibly modest impact.

Vol. 17, No. 4

ISSN 2064-7964

2023



Figure 2. The Oscillating Flow technique [7]

2.3. The pressure pulse decay technique

Brace developed the transient technique or pressure pulse decay method. The process for measuring permeability is as follows. As illustrated in Fig.3, the sample's input and output end faces were connected to the closed reservoirs while separated over the lateral surface. The fluid pressure in the sample and both tanks is the same and equal to P_0 at the beginning of the experiment. Then, a "pressure pulse" is given, and a stepwise pressure rises with a small value ΔP in the input reservoir. The fluid travels through the sample from the input reservoir into the output reservoir due to fluid pressure differences at the sample's input and output end faces. As a result of this motion, the fluid's pressure decreases in the input reservoir and increases in the output reservoir until they both achieve an equilibrium pressure P_A . Both the P_{in} and P_{out} reservoirs' pressure time dependences are noted [8].



Figure 3. The pressure pulse decay technique [8]

Suppose we have information on how the pressure changes over time in the input reservoir. In that case, it should be noted that pressure is theoretically easier to measure than the fluid volume, which should be done using the stationary approach. As a result, measures become more accurate. Because of this, the transient approach is unquestionably superior for measurements of materials with low permeability (on the order of 10^{-21} m² or even less).

Vol. 17, No. 4

ISSN 2064-7964

2023

3. HYDROGEN PERMEABILITY

Increasing the proportion of renewable energy in energy production requires large-scale energy storage. Power-to-gas technology may solve several problems with renewable energy sources erratic energy generation, which is now their biggest barrier to widespread adoption [9]. Power-to-gas systems use PEM electrolysis or methanation to transform surplus renewable energy into hydrogen gas or methane. This technique, which is already well established, may help to lower CO_2 emissions [10]. Natural gas networks can be filled with hydrogen, which can be done using the already-built infrastructure. Although considerably more difficult than storing natural gas, hydrogen storage is still a major challenge. When microorganisms are present, high levels of mobility, lightness, and reactivity have adverse impacts on hydrodynamics that are not minor and raise safety issues.

Nevertheless, there is already evidence for large-scale hydrogen storage [11]. The rock permeability, sealing characteristics, and general geomechanics of the cavern have all been extensively studied and modeled. To store natural gas, this method employs subterranean hard rocks. Fully cut off from the outside world is the reservoir. Hard rock serves only as a mechanical foundation where the cavern is dug. No isolating qualities are required for base rock. The vital installations, like drainage, are then performed, and the shotcrete reinforcement is added. The sealing layer comes last.

4. EQUIPMENT AND CONFIGURATION FOR EXPERIMENTS

Several designs can be used to study hydrogen. Dawid and Marcin David [12] studied hydrogen permeability using steady-state flow and carrier gas methods. Depending on the sample permeability, the setup between the two approaches indicated can be changed. The configuration is displayed in Fig. 4. The gas cylinder, pressure control valves, sample holder, accurate gas pressure transducers, and gas concentration detector make up this apparatus.



Figure 4. Setup for the steady-state and carrier gas methods [12]

DOI: https://doi.org/10.14232/analecta.2023.4.61-68

Vol. 17, No. 4

ISSN 2064-7964

On the upstream side, a reference gas of 10% hydrogen in methane was employed. The sample, contained by the water-constricting pressure of the PVC sleeve, is given gas. 1.0 MPa feed gas pressure was used during the tests. At a pressure of 100 kPa, carrier gas (helium) was injected into the downstream side of the device during the test. A single hydrogen gas detector with a sensitivity range of 2-2000 ppm was plugged into the setup's end for periodic gas concentration measurements. The same socket is used to plug in the carrier gas (helium) that the downstream side was filled with following vacuuming after the measurement is complete. When there is a flow of gas through the sample, a back pressure valve can be utilized to modify the pressure on the downstream side. A back pressure valve can maintain a constant pressure gradient across the sample by regulating the pressure on the upstream and downstream sides.

Hydrogen, methane, nitrogen gases, and sodium chloride were utilized in the studies by Amin et al. [13]. Two sandstone and one carbonate rock sample were collected from gas reserves and utilized for displacement testing. The buoyancy technique, based on the Archimedes principle, was used to measure the porosities of the samples. A fan oven housing was used to regulate the temperature, as shown in Fig. 5.





DOI: https://doi.org/10.14232/analecta.2023.4.61-68

Vol. 17, No. 4

ISSN 2064-7964

During flooding tests, a pressure gauge, the Rosemont 3051 pressure transmitter from Emerson, was utilized to constantly record the differential pressure across the sample (i.e., between the input and outflow faces of the core sample). The core samples were placed into the core holder and saturated with brine with the appropriate salinity after vacuum drying at 70 °C. Next, the brine was injected at three different flow rates to calculate K_w using Darcy's equation. For the calculations of relative permeability, the determined brine permeability served as the base fluid's (absolute) permeability. Finally, the appropriate gas (i.e., H₂, N₂, or CH₄) was introduced into the brine-saturated core sample at a predetermined constant pressure specified by the core's K_w value. Throughout the injection procedure, the input and outlet gas flow rates, the pressure difference between the core faces (i.e., inlet and outlet), and the volume of generated effluent brine were all recorded over time. Recording continued until no more brine was visible in the effluent.

Rock core encased in a Teflon—PTFE tube was employed by Yekta et al. [14], as illustrated in Fig. 6.



Figure 6. Setup for rock core encased in a Teflon—PTFE [14]

The measurement cell (core plus Teflon container tube) is then put within a cylindrical pressure vessel. The latter is pressured by injecting water using a hydraulic (Maximator) pump. During the measurements, the cell is subjected to continuous external pressure of 130 bar. This makes it possible to prevent any fluid movement between the core and the Teflon tube by maintaining the external confining pressure at a minimum of 30 bar above the fluid pressure within the cell. The vessel is placed inside a cylindrical furnace that an

Vol. 17, No. 4

Analecta Technica Szegedinensia

electronic regulator controls. An internal thermocouple situated just above the cell continuously measures the temperature within the vessel.

5. AUTOMATED PERMEABILITY MEASUREMENT

In practice, the permeability test of the rock sample is most often performed with equipment that works on the principle of pressure drop. This procedure can be automated very well, so the presence of a permanent person is not required during the measurement, and the measured data can be collected and evaluated easily. A schematic diagram of a system operating on the principle of pressure decay can be seen in Fig.4. The following markings can be seen in the diagram:

E-01 Filling tank, FP-01 Liquid pump, V-08 Pressure reducer, GP-01 Gas bottle, VP-01 Inlet buffer, VP-02 Outlet buffer, PT-02 Differential pressure transmitter, PT-01 Line pressure transmitter, TC-01 Digital temperature controller, TT-01 Temperature transmitter, TE-01 Air thermostat, E-02 Drain tank, CE-01 Rock core trap, VP-03 Oil/water separation tank, PT-03 mantle pressure transmitter, C-XX connection points, V-XX valves, DAQ-01 data collection/display computer, MP-01 mantle pressure pump.



Figure 7. schematic diagram of a system operating.

6. CONCLUSION

The focus of the application is the examination of the permeability of the reservoir rock. Based on the developed procedure, a new device is developed that is suitable for carrying out the measurements necessary to determine the permeability of the reservoir rock when using a hydrogen-methane mixture. With the help of the measurement results, it can be determined how the hydrogen-natural gas mixture affects the rock permeability of the used reservoir.

DOI: https://doi.org/10.14232/analecta.2023.4.61-68

Vol. 17, No. 4

ISSN 2064-7964

ACKNOWLEDGEMENTS

Project no. RRF-2.3.1-21-2022-00009, titled National Laboratory for Renewable Energy, has been implemented with the support provided by the Recovery and Resilience Facility of the European Union within the framework of Program Széchenyi Plan Plus.

REFERENCES

- [1] A. A. Pek, Dynamics of Juvenile Solutions (Nauka, Moscow, 1968).
- [2] W. S. Fyfe, N. J. Price, and A. B. Thompson, Fluids in the Earth's Crust (Elsevier, New York, 1976; Mir, Moscow, 1981).
- [3] W. F. Brace, "Permeability of Crystalline and Argillaceous Rocks," Int. J. Rock Mech. Mining Sci. Geomech. Abstracts 17 (5), 241–251 (1980).
- [4] V. M. Shmonov, V. M. Vitovtova, and A. V. Zharikov, Fluid Permeability of Crustal Rocks (Nauchnyi Mir, Moscow, 2002).
- [5] A. V. Zharikov, V. I. Mal'kovsky, V. M. Shmonov, et al., "A Method for Measuring the Permeability of Rocks Samples Including Changes in Thermodynamic Properties of the Fluid," Elektronnyi Nauchno-Inform. Zh. "Vestnik Otdeleniya Nauk o Zemle RAN", No. 1(22 (2004)
- [6] J. Bear, D. Zaslavsky, and S. Irmay, Physical Principles of Water Percolation and Seepage (UNESCO, Paris, 1968).
- [7] R. L. Kranz, J. S. Saltzman, and J. D. Blacic, "Hydraulic Diffusivity Measurements on Laboratory Rock Samples Using an Oscillating Pore Pressure Method," Int. J. Rock Mech. Mining Sci. Geomech. Abstracts 27 (5), 345–352 (1990).
- [8] G. J. Fischer and M. S. Paterson, "Permeability and Storage Capacity during Deformation at Elevated Temperatures," in Fault Mechanics and Transport Properties of Rocks (Academic, St. Diego, 1992), pp. 187–211.
- [9] Paul, D.; Ela, E.; Kirby, B.; Milligan, M. The Role of Energy Storage with Renewable Electricity Generation; National Renewable Energy Laboratory: Golden, CO, USA, 2010.
- [10] Bailera, M.; Lisbona, P.; Romeo, L.M.; Espatolero, S. Power to Gas projects review: Lab, pilot and demo plants for storing renewable energy and CO2. Renew. Sustain. Energy Rev. 2017, 69, 292–312.
- [11] Panfilov, M. Underground and pipeline hydrogen storage. In Compendium of Hydrogen Energy; Elsevier BV: Amsterdam, The Netherlands, 2016; pp. 91–115.
- [12] Gajda, D.; Lutyński, M, "Hydrogen Permeability of Epoxy Composites as Liners in Lined Rock Caverns—Experimental Study," Appl. Sci. 2021, 11, 3885. <u>https://doi.org/10.3390/app11093885</u>
- [13] Rezaei, A., Hassanpouryouzband, A., Molnar, I., Derikvand, Z., Haszeldine, R. S., & Edlmann, K. (2022). Relative permeability of hydrogen and aqueous brines in sandstones and carbonates at reservoir conditions. Geophysical Research Letters, 49, e2022GL099433. <u>https://doi.org/10.1029/2022GL099433</u>
- [14] Yekta, A.E., Manceau, JC., Gaboreau, S. *et al.* Determination of Hydrogen–Water Relative Permeability and Capillary Pressure in Sandstone: Application to Underground Hydrogen Injection in Sedimentary Formations. *Transp Porous Med* **122**, 333–356 (2018). https://doi.org/10.1007/s11242-018-1004-7

DOI: https://doi.org/10.14232/analecta.2023.4.61-68