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AN EVALUATION OF HYDROCARBON DEPOSIT TIGHTNESS IN ASPECT OF CO₂ SEQUESTRATION**

1.   INTRODUCTION

The origin of a hydrocarbon deposit is inseparably connected with the existence of a geological trap formed by reservoir rocks and sealing rocks, the shape of which favors the accumulation of hydrocarbons. Reservoir rocks have a number of features facilitating such accumulation and also the filtration of formation fluids. Reservoir rocks have free spaces in their structure in the form of pores between grains or inside them, micro-caverns or fractures, where oil and/or natural gas accumulate. These free spaces have to be interconnected to enhance the migration of hydrocarbons and water. The basic element of the trap is the sealing rock lying directly above the reservoir rock. Sealing rocks are essential for the formation of a natural reservoir, and so of a deposit, too. Characteristically such rocks have very low permeability and plasticity. Among sealing rocks we have, e.g. clays, clayey shales and evaporates. Hydrocarbons accumulate in the traps under the influence of a capillary barrier closing migration pathways for hydrocarbons. The tightness of this barrier depends on the type and thickness of the caprock as well as the lithological type of the rocks [14].

From a hydrodynamical point of view no ideally tight deposits exist. Escapes of hydrocarbons are common which is confirmed by self-spills of oil and gas or emissions of hydrocarbons from bituminous sands, deposits of asphalt oil. Most of the first classic oil and natural gas deposits were discovered in this way. Hydrocarbons may escape from deposits through the following pathways:
– damaged wells – loss of tightness caused by the corrosion of pipes, cement corrosion or incorrect cementing,
– faults and natural systems of fractures,

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- migration through the caprock (capillary forces were overcome),
- mechanical fractures (exceeded fracturing pressure),
- diffusion of the caprock.

The Darcy flow is a dominant mechanism of transport and migration of hydrocarbons in a porous rock matrix, as well as in a network of fractures and cracks. In the case of one-phase flow in a porous medium the absolute permeability is established between the flow rate of a fluid of a given viscosity and the existing gradient of pressure. When the rock is saturated with two or more immiscible fluids (oil, gas, water) a multi-phase flow will be observed in the porous medium, with the accompanying capillary effects. Then, the transport of fluid will depend on the surface tension on the contact of the displacing phases, the wettability of the rock (angle of wettability) and structure of the pore system.

2.   INTEGRITY AND TIGHTNESS OF THE CAPROCK

The caprock is indispensable and yet a most poorly recognized geological element of hydrocarbon deposits. It forms a barrier for the migrating hydrocarbons in a porous medium. Tight caprock is characterized by sufficient thickness, lateral continuity, plasticity and well as low porosity and permeability.

Escapes of hydrocarbons through the overburden rocks are commonplace. Hydrocarbons may migrate from the deposits through the existing micropore canals or fractures in the caprock. When hydrocarbons escape through the micropores then we have to do with the membrane sealing. When the escape takes place through the fractures opened by reservoir pressure, then we speak about hydraulic sealing [11].

The analysis of the membrane tightness of the caprock lies in defining capillary pressure, i.e. pressure at which the membrane loses its tightness. Such analyses are usually performed on caprock samples with the mercury porosimetry methods in laboratory conditions. The hydraulic tightness of the caprock is determined by making tightness tests in wells, i.e. a pressure value is defined for which the hydraulic barrier becomes leaky.

Although the sealing rocks may be treated as tight for hydrocarbons, they are not completely impermeable. Two major mechanisms are related with the migration of hydrocarbons of the caprock: molecular diffusion through the water-saturated pore canals of sealing rocks and slow Darcy flow of compressible gaseous phase. Molecular diffusion is a slow process and can be analyzed only in the geologic time scale. The convective flow resulting from the Darcy law mainly depends on the geological and hydrodynamic conditions of the deposit/sealing rocks system, and also properties of fluids in the deposit and in the sealing rocks. A slow flow (according to Darcy law) takes place when the difference of pressures between the top and the bottom of the sealing rocks is sufficiently high to overcome the capillary forces at the interface of the wetting phase (brine), which saturates the sealing rocks, and non-wetting phase (gas, oil), which fill the pore space of the deposit.

3.   DIFFUSION TRANSPORT

Migration of hydrocarbons to and through the sealing rocks of the caprock is caused by the diffusion transport through a system of pores saturated with water. This process takes

place continuously and everywhere. However, due to its slow rate (as far as the deposit tightness is concerned), therefore can be analyzed only in the geologic time scale, i.e. tens of million of years. The propagation rate of the gas diffusion front through the sealing caprock can be evaluated for various values of effective molecular diffusion on the basis of the Fick equation. In the steady state the diffusion flux of mass is defined as:

$$J = -D_{eff} \cdot \frac{\Delta C}{\Delta x} \tag{1}$$

For the caprock of thickness h and porosity ϕ and gas concentration in the pore water at the deposit/rock boundary (C_{aqu}) and assumed zero gas concentration in the top of the sealing rocks, the diffusive flux of mass flowing in a steady state of gas can be defined as:

$$J = -D_{eff} \cdot \frac{\phi \cdot C_{aqu}}{h} \tag{2}$$

The magnitude of flux migrating through the sealing caprock will be a function of gas concentration between the bottom and the top of the sealing caprock and is proportionate to the effective diffusion coefficient. The effective diffusion coefficient for gas in water for low-permeable rocks equals to $10^{-10} - 10^{-12}$ m²/s [4].

If the effective diffusion coefficient is of 10^{-10} m²/s, the diffusive stream of carbon dioxide flowing through the caprock of porosity 10% and thickness 10, 100 and 1000 m at a depth of 2,000 m will be 1,640, 164 and 16.4 kg/m²/Ma (Ma – million years), respectively [4]. When carbon dioxide is injected to oil deposits (EOR, CO_2 sequestration), where no gas has been present before, some lag-time should be accounted for. In this way the steady state diffusive stream flowing through the sealing rocks will be obtained in a function of time. Figure 2 illustrates the time needed for obtaining a 90% diffusive steady stream in a function of the caprock thickness for the effective diffusion coefficient of $10^{-10} - 10^{-12}$ m²/s.
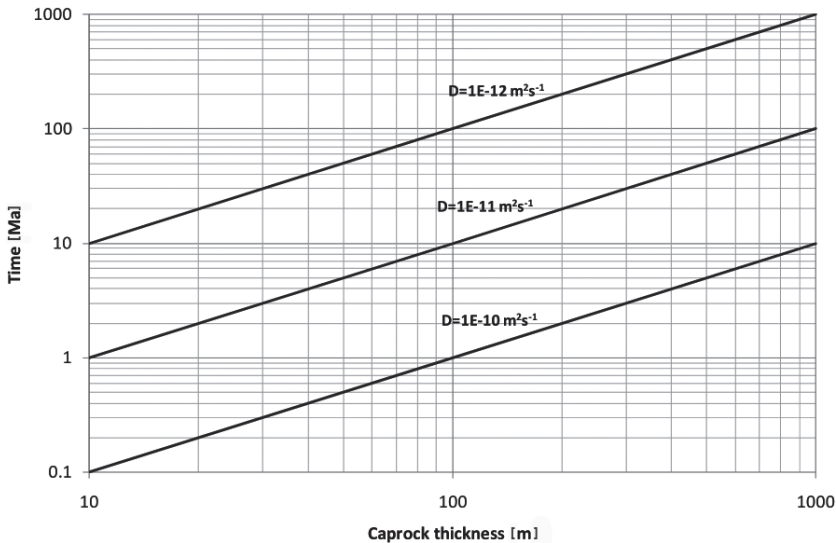

Fig. 1. Time needed for obtaining a 90% of the established diffusive stream in a function of caprock thickness [9]

The analysis of the plot in Figure 1 reveals that in the worst case (effective diffusion coefficient 10^{-10} m²/s and caprock thickness of 10 m) the time needed for obtaining steady conditions of diffusion gas stream is 0.1 mln years. Therefore, the escapes of carbon dioxide injected to the deposit as a result of gas diffusion for even such rock thicknesses are negligible. In the case of carbon dioxide the diffusion may also have an influence on some geochemical reactions, which in turn may result in changes of mechanical properties of sealing rocks.

4. MEMBRANE SEALING – MIGRATION THROUGH THE SEALING ROCKS

Generally, the sealing abilities of the caprock are determined by the minimum pressure needed for displacing water from the pore canals, thus opening the pathways for the migration of hydrocarbons through the top of the sealing rocks. Unlike the caprock thickness, the capillary pressure and the diameter of the canal connecting pores in the rock are important for the membrane sealing. In hydrostatic conditions only the buoyancy and capillary forces act on hydrocarbons in a water-saturated rock. The hydrostatic buoyancy force depends on the difference of water and hydrocarbon density. The bigger the density difference is, the higher is the buoyancy of a given column of hydrocarbons. On the other hand, the capillary forces are related with the wettability of the rock, the diameter of the contractions of the pore canals and surface tension on the hydrocarbon/water interface. The sealing ability of the caprock decreases with the increase of surface tension and lowers with the drop of rocks wettability and the decrease of the pore diameter. In reservoir conditions the parameters characterizing formation fluids, i.e. density, surface tension or parameters characterizing the porous medium, i.e. diameter of the pore contractions basically do not change, therefore are assumed to be constant. A variable parameter influencing the tightness of the caprock is the height of the hydrocarbons in the deposit. The critical height of the column provided by the membrane tightness describes the following dependence [3]:

$$Z_c = \left[2 \cdot \sigma \cdot \left(\frac{1}{r_t} - \frac{1}{r_p} \right) \right] \Big/ \left[g \cdot (\rho_w - \rho_o) \right] \qquad (3)$$

$$r_t = \left[\frac{1}{2} \cdot (0.154 \cdot D) \right]$$

$$r_p = \left[\frac{1}{2} \cdot (0.414 \cdot D) \right]$$

where:
H_c – critical height,
σ – surface tension,
r_t – radius of pore contraction,
r_p – pore radius,
D – grain diameter,
g – acceleration of gravity,
σ_w – water density,
σ_o – oil density

The graph presented in Figure 2 illustrates a dependance between the critical height of the oil or gas column as a function of grain diameter for two different fluid densities.
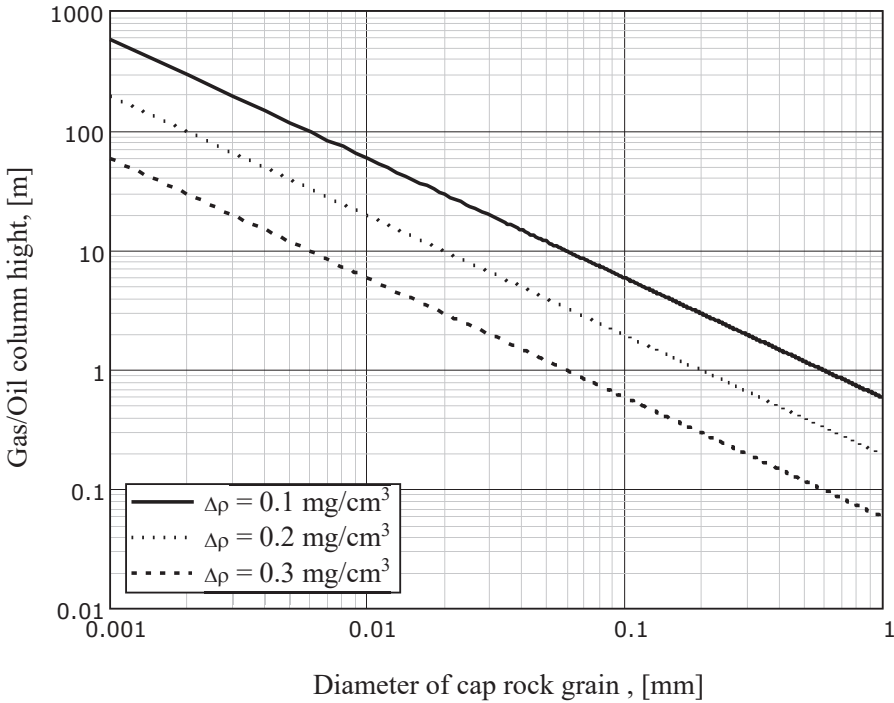

Fig. 2. Height of oil/gas column, which can be withheld by capillary forces in the overburden rocks of various grain diameter (surface tension σ = 35 dyna/cm)

The analysis of Figure 2 reveals that with the increase of grain size of the sealing rock and the concurrent increase of the density of water and hydrocarbons, the height of the column of hydrocarbons, which can be trapped by capillary forces acting in the overburden, will decrease. For instance, in a rock of 0.01 mm grain diameter and the assumed surface tension of 35 dyna/cm, difference of water and hydrocarbons of 0.1 g/cm³, the critical height of the oil column equals to 64.7 m. On the other hand, when the density gradient at the water/gas interface equals to 1 g/cm³, the height of the gas column equals to ca. 6.5 m.

5.   CAPILLARY PRESSURE

As already observed, the tightness of the water-saturated overburden rocks results from the existence of capillary pressure in the rock pores. When the pressure in the deposit exceeds the breakthrough pressure, at which a continuous flow of non-wetting phase takes place in the overburden capillaries, then hydrocarbons start to migrate though the sealing rocks and the deposit becomes leaky.

Capillary pressure defined as a difference of pressures between two immiscible fluids in a porous medium is described by the formula [1]:

$$P_c = P_{nw} - P_w = \frac{2 \cdot \sigma}{r} \cdot \cos\theta \qquad (4)$$

where:

P_{nw} – pressure of nonwetting phase,
P_w – pressure of wetting phase,
σ – interfacial tension,
θ – wetting angle,
r – radius of pore throat.

The analysis of equation (4) reveals that the breakthrough pressure is proportionate to the surface tension on the wetting/nonwetting fluids interface and inversely proportional to the radius of the pore canal. The angle of contact as a measure of rock wettability is another factor importantly influencing the capillary pressure value.

In Figure 3 we can see a pore canal with a curved surface of contact between nonwetting phase (hydrocarbons) and wetting phase (water), where P_n is a pressure in nonwetting phase, P_w is pressure of wetting phase and P_c is capillary pressure between phases in the pore canal. In this case the capillary pressure counteracts filtration of nonwetting phase to the rock mass.
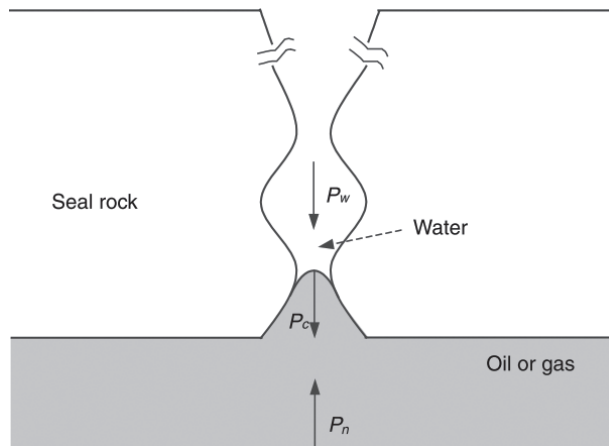

Fig. 3. Schematic of sealing mechanism in a single pore canal of a sealing rock [10]

When the difference of pressures between the nonwetting and wetting phases exceeds the capillary pressure in a given pore canal, i.e. $P_n - P_w > P_c$, the nonwetting fluid will move along the canal until it reaches another pore contraction of smaller diameter. When the difference of pressures in the sealing rock cross section exceeds the capillary pressure of a series of interconnected canals, then a continuous stream of nonwetting phase is formed and (according to Darcy law) nonwetting phase flows through the sealing rock. This gradient of pressures is a breakthrough pressure $P_{c,breakthrough}$.

Mechanism of hydrocarbon migration through non-homogeneous rocks of the overburden

The process in which a nonwetting phase (hydrocarbons) breaks through a water--saturated on-lying sealing rocks can be divided into stages, with the corresponding pressure of the nonwetting fluid. At the initial stage, when the nonwetting fluid pressure increases above the capillary entry pressure $P_{c,entry}$, the rock is partly saturated with this fluid. The capillary entry pressure, is a pressure above which the wetting phase is displaced from the unconnected pore canals of the largest diameter at the interface [2]. Exceeding of this characteristic pressure value allows migration of the nonwetting phase to the system of pores in the sealing rock, though the nonwetting fluid is not observed to flow yet. The further increase of pressure of the nonwetting fluid causes the opening of the successive smaller pore canals, displacement of wetting phase and an increase of saturation the nonwetting fluid in the pore system. At this stage the gas pressure is higher than the capillary entry pressure, but still lower than the breakthrough pressure ($P_{c,entry} < P_c < P_{c,breakthrough}$). When the nonwetting phase pressure increases above the breakthrough pressure the wetting phase will be displaced from the porous space to such an extent that interconnected flow pathways can be formed in the entire cross-section of the seal rock [5]. The interconnected pathways of the nonwetting fluid coincide with pore canals of largest diameter, where the capillary displacement resistance of phases is smallest. At this stage the flow of the nonwetting phase will cover only a small part of the interconnected pores. With a further increase of pressure new pathways are formed, increasing the saturation and effective permeability of the nonwetting phase. As a consequence the character of flow changes from capillary to viscous (Darcy) flow. The breakthrough pressure is an important parameter when assessing the sealing ability of rocks above the hydrocarbon deposit. Attention should be paid to the difference between capillary entry pressure and breakthrough pressure. The former is a measure of the diameter of the largest pore on the surface of the rock sample in given wettability conditions and the surface tension of fluids. The latter defined conditions in which a constant flow of hydrocarbons takes place through the on-lying sealing rocks [10].

After a gas breakthrough due to the existing difference of pressures we have to do with a continuous gas flow (the nonwetting phase), which is a function of effective permeability for this phase. The effective permeability of a rock is defined on caprock samples in a laboratory. During measurement the effective permeability for gas changes in time and is a function of saturation with a nonwetting phase (water). The measurement lies in recording changes of pressure on the sample outlet in a function of time. It is determined on the basis of the Darcy law from the equation [5]:

$$k_{eff} = -\frac{V_2 \cdot \mu \cdot 2 \cdot \Delta x}{A \cdot \left(P_2^2 - P_1^2\right)} \cdot \frac{dP_2}{dt} \tag{5}$$

where:

$\quad\quad V_2$ – volume of reservoir on the outlet of sample,
$\quad\quad P_1$ – pressure on the inlet of sample,
$\quad\quad P_2$ – pressure on the outlet of sample,
$\quad\quad A$ – surface of cross-section of sample,
$\quad\quad \mu$ – dynamic viscosity of gas,
$\quad\quad \Delta x$ – length of sample.

The lowering of the pressure of the nonwetting phase after the gas breakthrough will lead to the re-imbibition of the rock with the wetting phase, starting from pores of smallest diameter to pores of larger diameters. Gradual closing of canals of gas flow results in lowered effective permeability of the nonwetting phase. Finally, the last interconnected pore canal is closed and the gas flow is ultimately stopped. As a result of the lost connection between the pore canals saturated with the nonwetting phase the difference of pressure between gaseous phase under and above the sealing rocks increases. Its absolute value is a measure of the biggest and most efficient pore canal radius (r_{eff}), which also determines the efficiency of the capillary sealing of the rock. In the process of imbibition with the wetting phase part of the gas-saturated pores may be cut off as a result of damaged pathways for gas transport, which may result in the residual saturation with the nonwetting phase (gas).

Measurements of capillary tightness of sealing caprock are usually performed on rock samples taken from wells in laboratory conditions. Among the basic methods of determining capillary pressure belongs the mercury porosimetry method. During the tests mercury is injected to the air-saturated sample and then a capillary displacement plot is determined. Next, the plot is converted to the water/gas system and on this basis the breakthrough pressure is defined for gas [3]. However, the pressure at which the displacement begins corresponds more to the capillary entry pressure, which is much lower than the breakthrough pressure [7, 10].

The direct measurement of breakthrough pressure of sealing caprock is realized in the displacement of the wetting phase (water, brine) with the nonwetting phase (gas). In the course of this measurement the pressure of the nonwetting phase is gradually increased (in small steps). The breakthrough pressure is determined when a continuous, slow outflow of water is observed from the analyzed rock sample, followed by an outflow of nonwetting phase (gas). The direct measurement of breakthrough pressure of low-permeable sealing rocks is very time-consuming, therefore rarely performed. On the other hand it is the most reliable test.

6.   MEMBRANE TIGHTNESS OF ROCKS IN THE PRESENCE OF CO$_2$

When carbon dioxide is injected to the depleted oil and gas deposits while performing sequestration or secondary production, the overburden has to be assessed for tightness in the presence of CO$_2$. For the sake of preventing CO$_2$ escapes through the sealing overburden it is necessary to determine such a value of maximum injection pressure that the difference of pressures in the sealing rocks was lower than the breakthrough pressure. Otherwise the injected CO$_2$ will penetrate the sealing overburden and flow to the on-lying rocks. Most of the available literature data on the breakthrough pressure values refers to hydrocarbons/water system. These data cannot be directly used for assessing the breakthrough pressure in the CO$_2$/water system because of considerable changes in surface tension on the phase interface. Besides the wettability angle of the CO$_2$/water/rock system differs from that of hydrocarbons/water/rock system [10].

In the case of natural gas and oil deposits with gaseous cap, hydrocarbons are stopped in the deposit by the capillary forces on the gas/water interface in the sealing rocks. For oil deposits without the gaseous cap the breakthrough pressure for a given type of sealing rocks is determined by capillary pressure on the oil/water contact. In reservoir conditions, at

appropriately high pressure and temperature the surface tension at the oil/water interface is of 30 to 50 mN/m [3], so a few times higher than for the carbon dioxide/brine system. Therefore, the breakthrough pressure for CO_2 and sealing rocks will be lower than for oil. Sealing rocks, which are a barrier for the migration of hydrocarbons for oil and gas deposits may be insufficiently tight for CO_2 and allow its migration to the on-lying formations. The comparison of breakthrough pressure in the methane/brine and carbon dioxide/brine systems is presented in Figure 4. The breakthrough pressure for sealing rocks in the hydrocarbons/water system turns out to be much higher than for the CO_2/water system. The laboratory analyses of sealing rock samples of the overburden reveal that they differ in tightness depending on gases and increase $CO_2 < CH_4 < N_2$, being inversely correlated with the effective permeability of rock for these gases [4, 5, 10].
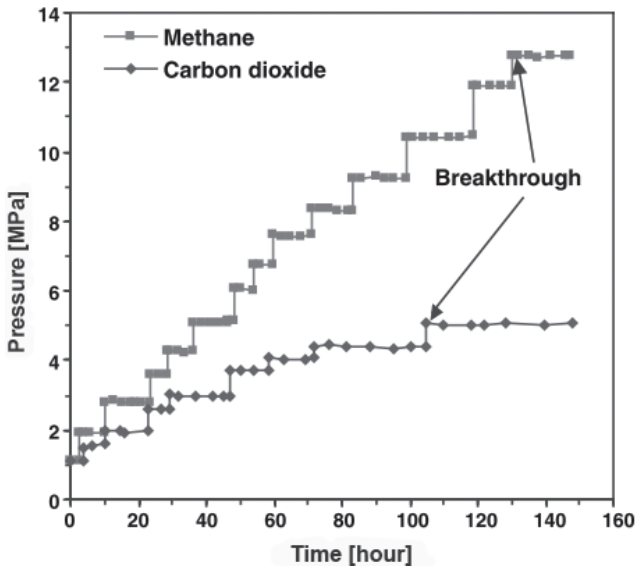

Fig. 4. Breakthrough pressure of sealing rock samples
in the methane/brine and carbon dioxide/brine systems [10]

The breakthrough pressure values may vary in a vast range from 0.1 MPa to 10 MPa at maximum admissible effective permeability for gas 10^{-18} to 10^{-24} m^2 (1,000 to 0.001 nD) [4].

It follows from the previous analysis that after the breakthrough pressure is exceeded, a slow and continuous flow of the nonwetting phase takes place in the sealing rocks. Basing on the dependence (5) we can determine the time after which gas breaks through the sealing rocks of a given thickness and of effective permeability. In fig. 5 we have a diagram showing the breakthrough time for CO_2 in the sealing rocks after exceeding the breakthrough pressure and admissible effective permeability from 1,000 to 0.001 nD and thickness from 10 to 1,000 m. An exemplary deposit located at 2,000 m, which corresponds to reservoir pressure of 20 MPa and temperature ca. 70°C, was analyzed. The assumed excess of reservoir pressure, which initiated the capillary transport of the nonwetting phase was ca. 2 MPa. The effective porosity of the sealing rocks was 1%. The analysis of the plot in fig. 6 reveals that in the

case of sealing rocks of relatively high permeability (ca. 1,000 nD) and thickness (10 m) the breakthrough time for carbon dioxide does not exceed one year. On the other hand, for the sealing rocks characterized by low permeability and thickness of 100 m the breakthrough time for carbon dioxide is of hundreds or even thousands of years, to increase with the growing thickness and drop of permeability of the caprock.
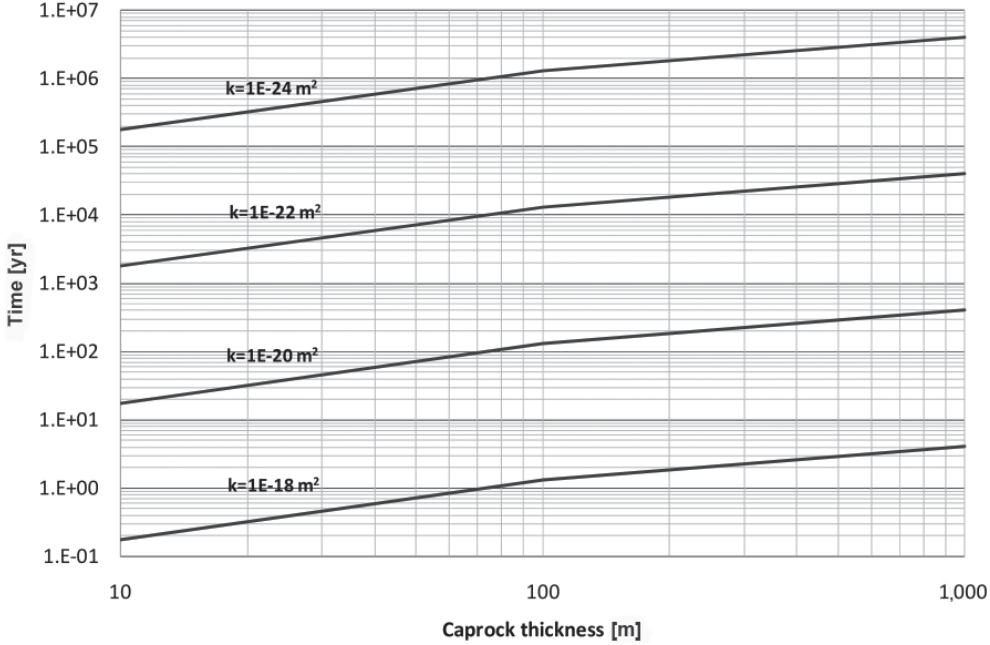

Fig. 5. Time of CO_2 breakthrough in the caprock after exceeding breakthrough pressure in a function of thickness and effective permeability of rocks [4]

The tightness of the sealing rocks is also influenced by the mutual interaction of the rock, water and carbon dioxide. The injected carbon dioxide contacts with the reservoir water forming carbonic acid, which will affect the permeability of the rock matrix. The existing experimental data indicate that these changes are slow and hard to quantitatively evaluate in time-limited laboratory conditions. Nonetheless, the kinetics of the process is low and its influence on the tightness of the overburden small [4].

7. GAS ESCAPES THROUGH FAULTS AND FRACTURES

Faults and fractures are considered to be potential pathways for gas escapes from deposit, limiting the structural integrity of traps and also deposits. The causes of this phenomenon may be various. The rocks may lose their tightness due to the primary fracturing of the intact caprock, being a consequence of reactivation of the existing faults or propagation of the existing faults in the rock matrix due to tangential or normal stresses in the fault [12].

A few mechanisms responsible for the sealing of throw planes, i.e. faults, can be distinguished [6, 13]:

– juxtaposition of rocks considerably differing in permeability, as a consequence of which highly permeable rocks (sandstone) contact with very low-permeable rocks (shales), which have high entry capillary pressure,

– clay smearing of fault surfaces, as a result of which the entry capillary pressure of the fault increases,

– cataclasis – sand grains are crushed, as a consequence of which the zones filled with fine material of high entry capillary pressure are formed,

– diagenesis, as a result of which selective cementing takes place along the fault surface. This may lead to partial or complete vanishing of porosity, and so hydraulic sealing of the fault.

Sealing of the deposit caused by a fault stems results here from different sealing properties of the contacting rocks. If the thrown reservoir rock contacts a compact, low-permeable rock (shale), then the sealing will be effective. Permeable rocks, e.g. sandstones which differ in capillary properties may also act as sealing. They differ in capillary properties, including difference of pressures in the fault surface, thanks to which the hydrocarbons can be stopped. The height of the oil column in this type of sealing may reach 15 m [3]. While evaluating sealing properties resulting from such effects as cataclasis, diagenesis or clay smearing we need to refer these mechanisms to measurable properties, e.g. lithology of rocks or fault displacement [13].

Escapes of hydrocarbons through this type of leaks are difficult to assess in laboratory conditions. This type of leaks are evaluated with permeability measurement methods for rocks in the entire deposit, e.g. water injection or leak-off tests.

8. CONCLUDING REMARKS

The formation of hydrocarbon deposits is connected with the presence of sealing rocks of appropriate shape, configuration and properties, which stop the further migration of hydrocarbons in the porous medium. Such rocks have very low permeability and sufficiently high thickness and plasticity.

The tightness of the overburden rocks is connected with two major mechanisms, i.e. membrane and hydraulic sealing. Membrane sealing is connected with the existence of capillary pressure in water-saturated pore canals of sealing rocks.

Capillary forces are related to rock wettability, the diameter of pore contractions and surface tension on the hydrocarbons/water interface. The sealing ability of the overburden increases with the growing surface tension and lowers with the decreasing wettability of rocks and decreasing diameter of pore contractions.

The sealing ability of the overburden rocks for hydrocarbons may be insufficient if carbon dioxide is injected to the deposit, as the capillary breakthrough pressure for CO_2 is smaller. Therefore, when carbon dioxide is to be stored in depleted workings or used in carbon dioxide-based EOR methods it is necessary to determine the breakthrough pressure for this gas.

Breaking of gas through the compact and water-saturated rocks of the overburden due to the operation of capillary forces may turn out impossible and the integrity of the overburden will be disturbed by the hydraulic leak of the caprock. In this situation the hydrocarbons migrate through the existing system of fractures, cracks or faults. This process is initiated mainly by increased reservoir pressure above its initial value or due to the tectonic movements.

Hydrocarbons also migrate through the caprock due to the diffusion, though this process is slow and is significant in the geologic time perspective. In the case of carbon dioxide the diffusion may influence some geochemical reactions, leading to the change of mechanical properties of the sealing rocks.

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