The sealing efficiency of cap rocks – laboratory tests and an empirical correlation

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The sealing efficiency of cap rocks – laboratory tests and an empirical correlation

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One of the major concern for the gas sequestration/storage feasibility in natural underground formations is the assessment of the sealing efficiency of the low-permeable sequences overlying potential storage formations. The sealing efficiency is quantified via the threshold pressure and/or residual pressure difference parameters; the experimental laboratory procedures for their evaluation have been widely investigate by the oil and gas industry at least from the fifteens of the last century. In order to evaluate the effect of adopted lab procedures, the present paper collects, categorizes and analyzes experimental data from the technical literature, investigating representative lithologies, via N2 brine fluid system, in reservoir thermodynamic condition. The analyses show an satisfactory coherence and consistency of data for a range of permeability of [10^2-10^5] nDarcy, independently from the adopted lab procedures. Data scattering for higher or lower permeability values seems to be more case sensitive rather than attributed to the adopted procedure or the investigated lithology. Furthermore, the empirical correlation by Thomas et al. (1968) was verified on the bases of the collected data. Even if the equation was derived only from standard test measurements on a limited number of lithologies, it turns out to be a reliable predictive approach for the identification, at least, of the order of magnitude of the threshold pressure (or the residual pressure difference) if only absolute permeability values are availlable.

Keywords: *sealing efficiency, fine-grained lithology, displacement pressure or residual pressure difference, breakthrough or threshold pressure, drainage-imbibition process, capillary processes.*

1. Introduction

The storage of materials in solid, liquid or gaseous phase into natural underground formations, such as aquifers, depleted hydrocarbon reservoirs, salt caverns, mines, has been widely adopted all over the world targeting different society needs: from the waste (radioactive and nonradioactive) sequestration to the cyclical and seasonal injection and production of natural gas for suppling daily and seasonal energy demand (Coti *et al.*, 2018; Benetatos *et al.*, 2020). Furthermore, the gas storage or sequestration in natural formations is supposed to play a crucial role in the energy transition process as well as in mitigating the anthropogenic emissions of greenhouses (Benetatos *et al.*, 2019). Great attention has been focus on the geological sequestration of CO_2 as well as on the large-scale storage of energy from renewable or green source (Wollenwebet *et al.* 2010, Bocchini *et al.*, 2017). Porous sedimentary formations hosting hydrocarbon reservoirs or aquifers represent the best candidates due to their high storage capacity, their elevate numbers and distribution worldwide and, regarding the depleted hydrocarbon reservoirs, due to all the available facilities and the high "confidence" with the system acquired although the reservoir production life (Verga, 2018).

One of the major concern for the gas sequestration/storage feasibility is the assessment of the sealing efficiency of the low-permeable sequences (denominated caprock) overlying potential storage formations (Wollenwebet *et al.*, 2010). These fine-grained pelitic rocks (i.e.: clay, claystone, shale, mudstone, siltstone) act like flow barriers avoiding gas migration from the reservoir to the adjacent upper formations. It is not trivial to remind that the accumulation of hydrocarbon reserves is guarantee by the presence of the caprocks: acting like a trap, they hinder the vertical migration of hydrocarbons due to buoyancy forces (Hantschel and Kauerauf, 2009).

The main phenomena potentially compromising the caprock sealing efficiency are: the induced mechanical fractures, the molecular diffusion through the water-saturated pore space of the seal formation and the slow Darcy flow of a free gas phase through the formation (Li *et al.*, 2005). The last is the focus of the present paper. The Darcy flow is induced if the capillary pressure increment due to gas injection in the reservoir overcomes the caprock sealing capacity, which standardly is express in term of threshold pressure and/or residual pressure difference. Both parameters are capillary pressure and they are estimated via dedicated laboratory analyses. Furthermore, because the sealing efficiency of pelitic formations impacts directly on the volume and location of hydrocarbon accumulation, defining potential exploration targets, (Egermann *et al.*, 2006), these parameters as well as their experimental determination have been widely investigated by the oil and gas industry at least from the fifteens of the last century. An extensive panorama is available in the technical literature (Wyllie and Rose, 1950; Thomas *et al.*, 1968; Schowalter, 1979; Hildenbrand *et* *al.*, 2002; Li *et al.*, 2005; Egermann *et al.*, 2006; Ito *et al.*, 2011). Nevertheless, a clear and straightforward analysis of it is not a trivial task, starting from the coherence and the definition of the adopted terminologies and nomenclature, until the developed lab test procedures, with drawbacks and advantages. The present paper collects, categorizes and analyzes experimental threshold pressure and/or residual pressure data availlable in the technical literature. The performed analyses focus on the effects of differ experimental procedures on the sealing efficiency results. Furthermore, the reliability of the threshold pressure prediction via an empirical correlation defined by previous research was evaluated.

2. Theoretical background: capillary pressure, drainage and imbibition processes

The capillary pressure, *Pc*, is the pressure difference between two immiscible phases. It is defined as the difference between the pressure of the non-wetting fluid (usually the hydrocarbon) and the pressure of wetting fluid (usually the brine) in the pore throats of a porous media. The capillary pressure, *Pc*, in a pore throat of equivalent radius, *r*, is expressed by the Washburn equation (Washburn, 1921):

$$P_C = \frac{2\sigma \cos\theta}{r} \qquad (1)$$

where σ is the interfacial tension (IFT) between the non-wetting phase and wetting phase and θ is the contact angle of the pore surface. The capillary pressure is a function of: the structure of the pore system, the wettability of the solid with respect to the fluid, the characteristic of the immiscible fluids and the thermodynamic condition of the system (through the dependence of σ and θ on pressure and temperature).

Concerning the drainage/re-imbibition phenomena (fig. 1), if the gas pressure apply to a pelitic formation 100% sutured by brine is sufficiently high to induce an entry pressure, $P_{c,\ entry}$, overcoming the sealing capacity of the formation, the non-wetting gas phase will start to enter in the pore throats displacing the wetting brine phase. The drainage process starts. At this stage, the gas flow is concentrated in a small portion of the largest interconnected pores because they provide the least resistance to capillary forces. If the initial gas pressure is enough high or if its increment continues further, additional fluid flow pathway will evolve in the porous medium with increasing divergent and the flow regime will change form capillary dominated to viscous dominated (Hildenbrand *et al.*, 2002). At this new stage the breakthrough pressure, $P_{c,\ breakthrough}$ or threshold pressure, $P_{c,\ threshold}$, is reach. Quoting form Hildenbrand *et al.* (2002): "This term refers to the excess pressure in the non-wetting phase at which the wetting phase is displaced to an extent that the percolation threshold is exceeded and continuous flow-paths of non-wetting phase form across the pore system". In the last stage, after the gas breakthrough is occurred and the gas excess pressure is reduced, a spontaneous re-imbibition process occurs: the brine (wetting phase) displaces back the gas starting from the smallest pores. The imbibition process is characterized by a constant and continuous decrease of the gas saturation, until a minimum (but not null) value is reach. It corresponds to the non-interconnected gas volume trapped by the capillary force. Consequently, a pressure difference persists between the gas phases below and above the seal plug, the so called minimum displacement pressure P_d, or residual pressure difference, $P_{c,\ residual}$ (Hildebrand *et al.*, 2002).

During drainage and imbibition processes, both immiscible fluids are able to flow across the porous medium: two relative permeability curves (fig. 2) can be adopted to describe the flow of each phase according to its saturation and

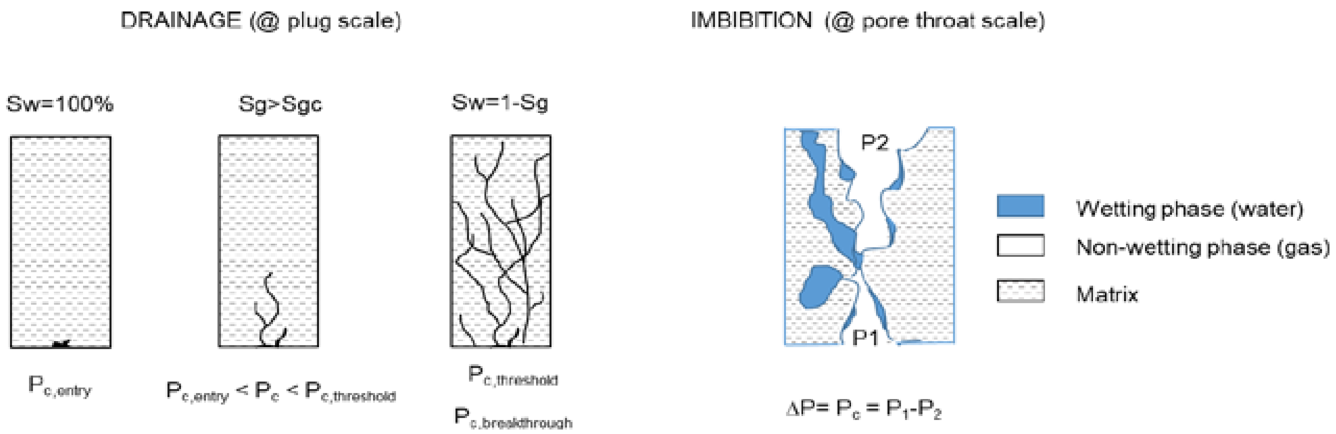

Fig. 1 – Drainage and re-imbibition processes in a fine-grain porous medium (modified by Hildenbrand *et al.*, 2002).

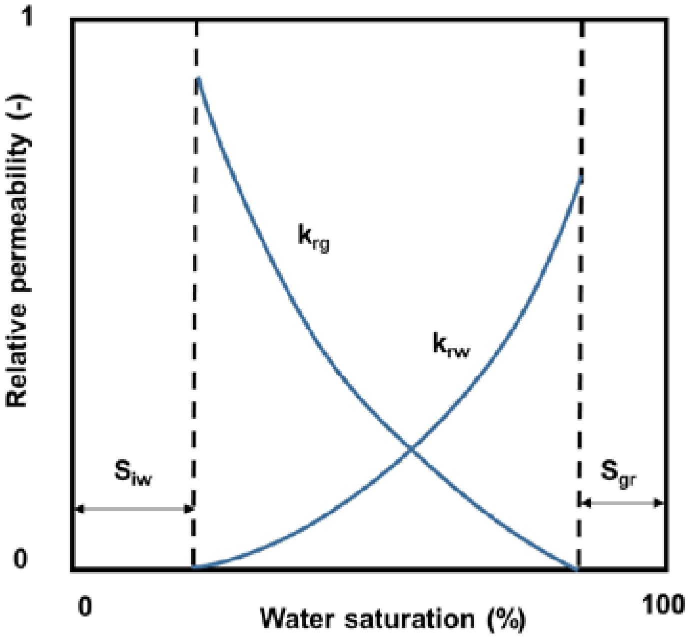

Fig. 2 – Example of relative permeability curves for a gas-water system (Ahmed, 1946).

the wettability of the system. The relative permeability is defined as the ration between effective and absolute permeability. The absolute permeability is a property of the porous medium and is a measure of the capacity of the medium to transmit fluids; the effective permeability is the conductivity of each phase at a specific saturation (Ahmed, 1946). In particular, if the gas is the non-wetting phase and the brine is the wetting phase initially fully saturating the porous media, during the drainage process the relative permeability of gas is zero until the critical gas saturation, S_{gc}, is reach. It is the minimum value of gas saturation at which the phase becomes movable. Increasing the gas saturation, its relative permeability increases as well until a maximum value is reach at the end of the drainage process. The following imbibition process results in a continuous reduction of the gas permeability because of the successive loss of interconnected flow-paths, which ultimately is shut off and the relative permeability of gas drops to zero. At the end of the imbibition phenomena, a residual gas saturation, S_{gr}, exists in the porous media. If a successive drainage process is induced in the same porous media, the residual gas saturation is already present and the gas flow will be benefit consequently.

3. *Laboratory tests for the evaluation of the caprock sealing efficiency*

In the last 60-70 years different laboratory procedures have been developed and fully described in the technical literature for the quantification of the sealing efficiency of a caprock. The simplest approach consists in deriving the *in-situ* threshold pressure form mercury porosimetry curve but the obtained results are quite always supported by the values obtained from the "standard or step-by-step approach" and/or the "residual capillary pressure approach" (according to Egermann *et al.* (2006) nomenclature). They are the most accredited and adopted procedures and they are based on the key idea of a drainage or drainage and re-imbibition test, respectively, developed at *in-situ* confining stress and temperature. As a common first step, a seal plug is fully saturated by brine and a single-phase fluid flow test is developed in steady state regime in order to evaluate the absolute permeability via the application of Darcy's law. According to the standard approach (Thomas *et al.*, 1968, Li *et al.* 2005, AL-Bazali *et al.*, 2005, among the others), a drainage test is successively developed: the brine in the seal plug is displaced by a non-wetting gas phase imposing a step-by-step increase of the inlet gas pressure. Each pressure step must be sufficient small not to overcome the expected threshold pressure and it is kept constant until the measured outlet brine flow is null. When the breakthrough pressure is reach, a continuous brine flow is monitored at the outlet face followed by an abrupt gas/liquid flow rate increment. Figure 3 exemplifies a standard test approach and interpretation according Li *et al.* (2005) paper. The standard approach is widely adopted for its accuracy and its robustness, nevertheless lab tests could become unacceptably time consuming: the rage of permeability of the seal plugs is in the order of mDarcy (10^{-15} m^2) to nDarcy (10^{-21} m^2) or even less. Furthermore, also a high accuracy in measuring very small flow rate is required. To mitigate these drawbacks, some variations of the test procedure was developed. As an example, Rudd *et al.* (1973) proposed a continuous injection of the non-wetting gas phase at a very small rate but the results seems to significantly overestimate the real threshold pressure values (Egermann *et al.*, 2006).

To overcome the limitation of the standard procedure related essentially to the experimental duration, Hildenbrand *et al.* (2002) proposed a test approach for the measurement of the residual capillary pressure at the end of a drainage and re-imbibition experiment. In summary, the initial inlet gas pressure applied at a

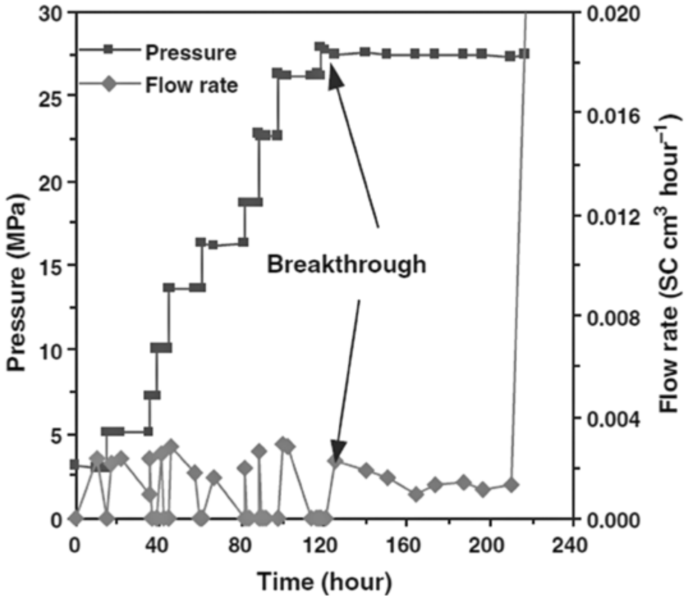

Fig. 3 – Results of a standard step-by-step approach by Li et al (2005)

brine saturated plug is higher than the expected threshold pressure; a drainage process is induced; the evolution of the pressure is monitored in the two closed upstream and downstream chambers, together with the temperature and the fluid volume. At the end of the successive and spontaneous re-imbibition process, a residual pressure difference persists between the two chambers due to capillary forces, the so-called minimum displacement pressure, P_d, or residual pressure difference, $P_{c,\ residual}$. The gas effective permeability curve can be computed via Darcy flow for compressible media (Ahmed, 1946). Figure 4 schematizes the two versions of the Hildenbrand *et al.* (2002) approach (a) together with experimental results (b) from Hildenbrand *et al.* (2004).

The standard procedure and the residual capillary pressure procedure quantify the sealing efficiency of the formation via two different parameters, which, at least, might coincide. Quoting Hildenbrand *et al.* (2002):"The residual pressure differences recorded at the end of the capillary-controlled, spontaneous re-imbibition process are interpreted to represent a good approximation of the threshold or breakthrough pressure from the drainage process. If re-imbibition is impeded, the residual imbibition pressure differences will be lower than the corresponding drainage threshold pressures. Comparison of our experimental results with literature data from drainage experiments suggest that this is likely for samples with permeabilities in excess of 100 nDarcy (10^{-19} m^2). Generally, imbibition data will result in conservative estimates (underestimation) of the capillary sealing efficiency".

Despite the experimental procedures, also the adopted fluid system plays a crucial role in the evaluation of the sealing capacity of a formation via lab tests. Even if the real phenomenon involves a sealing formation saturated by brine and a reservoir formation saturated by a gas mixture (usually hydrocarbon with substantially amount of nitrogen, sulfur, oxygen...), the lab experiments are usually performed with a nitrogen-brine fluid system, for sake of laboratory simplicity. Subsequently the results of N_2-brine system are converted to the hydrocarbon-brine system according to the following equation derived from the definition of capillary pressure:

$$\frac{P_c(gas-brine)}{P_c(N_2-brine)} = \frac{\sigma_{gas}\cos\theta_{gas}}{\sigma_{N_2}\cos\theta_{N_2}} \quad (2)$$

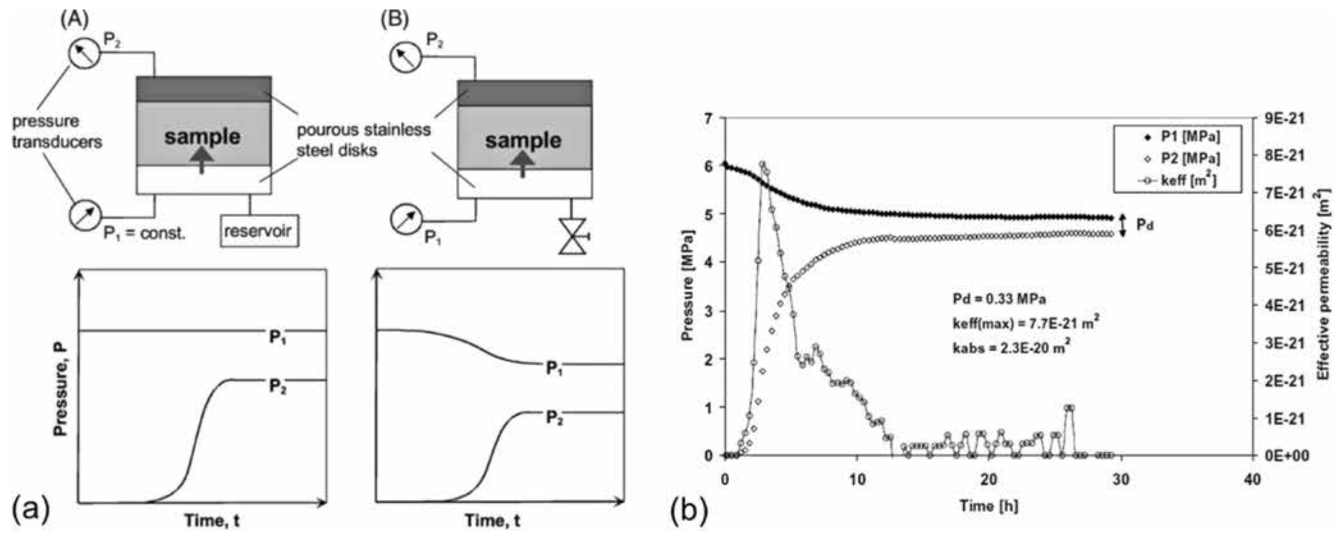

Fig. 5 – Residual capillary pressure procedure (a) and results of a lab test (b) (Hildenbrand et al, 2002 and (Hildenbrand et al, 2004).

Tab.1 – Sources of the dataset and classification according to the experimental procedures.

Standard approach	Residual capillary pressure approach	Standard + Residual capillary pressure approaches
Thomas *et al.*, 1968	Wyllie and Rose, 1950	Egermann *et al.*, 2006
Schowalter, 1979	Hildenbrand *et al.*, 2002	Ito *et al.*, 2011
Li *et al.*, 2005		

A reliable determination of the interfacial tension and the contact angle values is not a trivial task because of their dependency on the fluid system, on the characteristics of the porous medium (such as the wettability) and on the reservoir thermodynamic condition. Nevertheless, the correction via the equation (2) is widely adopted in the research papers, and the interfacial tension and contact angle values from literature is usually adopted in absence of direct lab evaluation. Finally, in particular and documented cases, the adoption of the *in-situ* fluid system for lab experiments is strongly recommended: for example, experimental evidences (Li *et al.*, 2005) have demonstrated that the interaction between CO_2 and the porous medium can alter its petrophysical properties, such as permeability, with consequent effects on the threshold or displacement pressure values. The research is still ongoing.

4. Data analysis and discussion

Several previous studies, experimentally investigating the sealing efficiency of sedimentary rocks, were collected and analyzed. They were categorized on the basis of the experimental procedures (table 1): the standard or step-by-step approach for the evaluation of the breakthrough pressure, $P_{c,\ breakthrough}$, or threshold pressure, $P_{c,\ threshold}$, and the residual capillary pressure approach for the minimum displacement pressure P_d, or residual pressure difference, $P_{c,\ residual}$. All the analyzed experiments were conducted at *in-situ* pressure and temperature conditions and adopting a N_2-brine fluid system.

The capillary pressure measures as a function of the absolute permeability were compared to appreciate the effect of the two different laboratory procedures, also taking into account the lithology. Furthermore, the dataset was use to confirm the reliability and the robustness of the empiric formula proposed by Thomas *et al.* (1968) for predicting the threshold pressure as a function of the solely absolute permeability:

$$P_{TH} = 7.37\left(\frac{1}{K}\right)^{0.43} \qquad (3)$$

where P_{TH} is the threshold pressure (*psi*) and K is the absolute permeability (mD). The formula was derived based on the results obtained by the standard approach experiment conducted adopting N_2-brine fluid system on 5 sandstone plugs, 2 limestone plugs and 1 dolomite plug. The investigated range of the absolute permeability was [40 – 1.78 10^5] nD, and the range of the measured threshold pressure was [0.06 – 5.17] MPa.

Figure 5 shows the comparison between all the collected lab results conducted with the two different approaches on the same plugs. The analyzed lithologies are: chalk, carbonate and sandstone (Egermann *et al.*, 2006); mudstone (Ito *et al.*, 2011). The consistency between the results and between the experimental data and the Thomas' equation is satisfactory: no clear effect of lithology can be appreciated. The discrepancy due to the experimental approaches seems to increase with increasing permeability; furthermore, also the mismatch between experimental data and the equation is maximum for the highest permeability values, which exceed the permeability range investigated by Thomas.

Figures 6 and 7 show the empirical equation together with all the collected data, categorized according to experimental procedures and to the source (fig. 6) and to the lithologies (fig. 7). The following consideration can be derived:

1. within its permeability range of definition, the empirical correlation is consists with the laboratory data from both procedures (i.e. breakthrough or threshold pressure values and the minimum displacement pressure or residual pressure difference values). The discrepancy increases for permeability lower then 10^1-10^2 nD and higher than 10^6 nD.
2. The discrepancy between the experimental results and between the experimental data and the Thomas' equation seems to be more case sensitive rather than attributed to the lab procedure or the investigated lithology. As discussed in the previous paragraphs, the lab results are influenced by the thermodynamic conditions (different for each test and not always specified), which affect the interfacial tension and the contact angle. Furthermore, as pointed out by the numerous revised papers, one of the crucial aspect is an accurate and reliable measurement of the flow rate and the pressure variation, both very

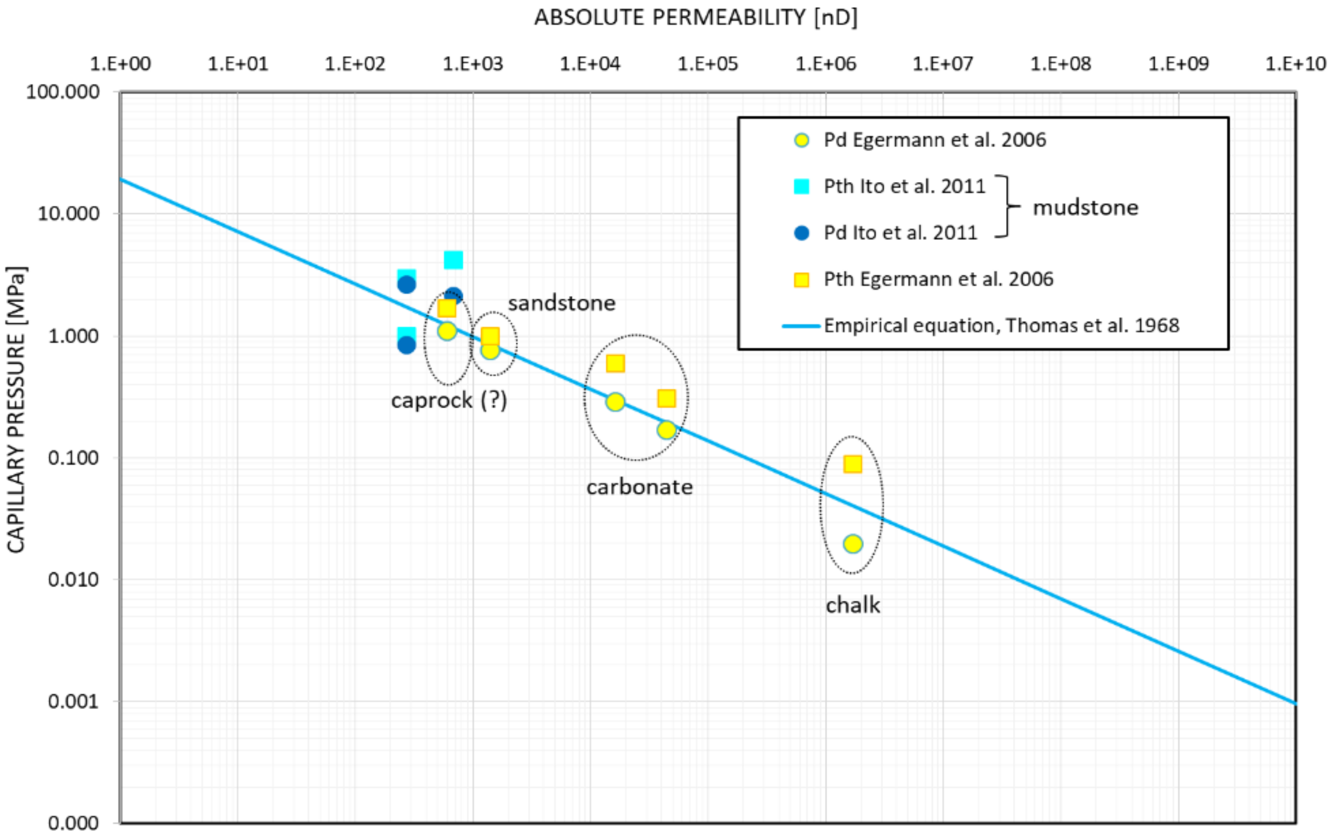

Fig. 5 – Comparison between the results from the standard and from the residual procedures performed on the same plug; Thomas *et al.* (1968) empirical equation.

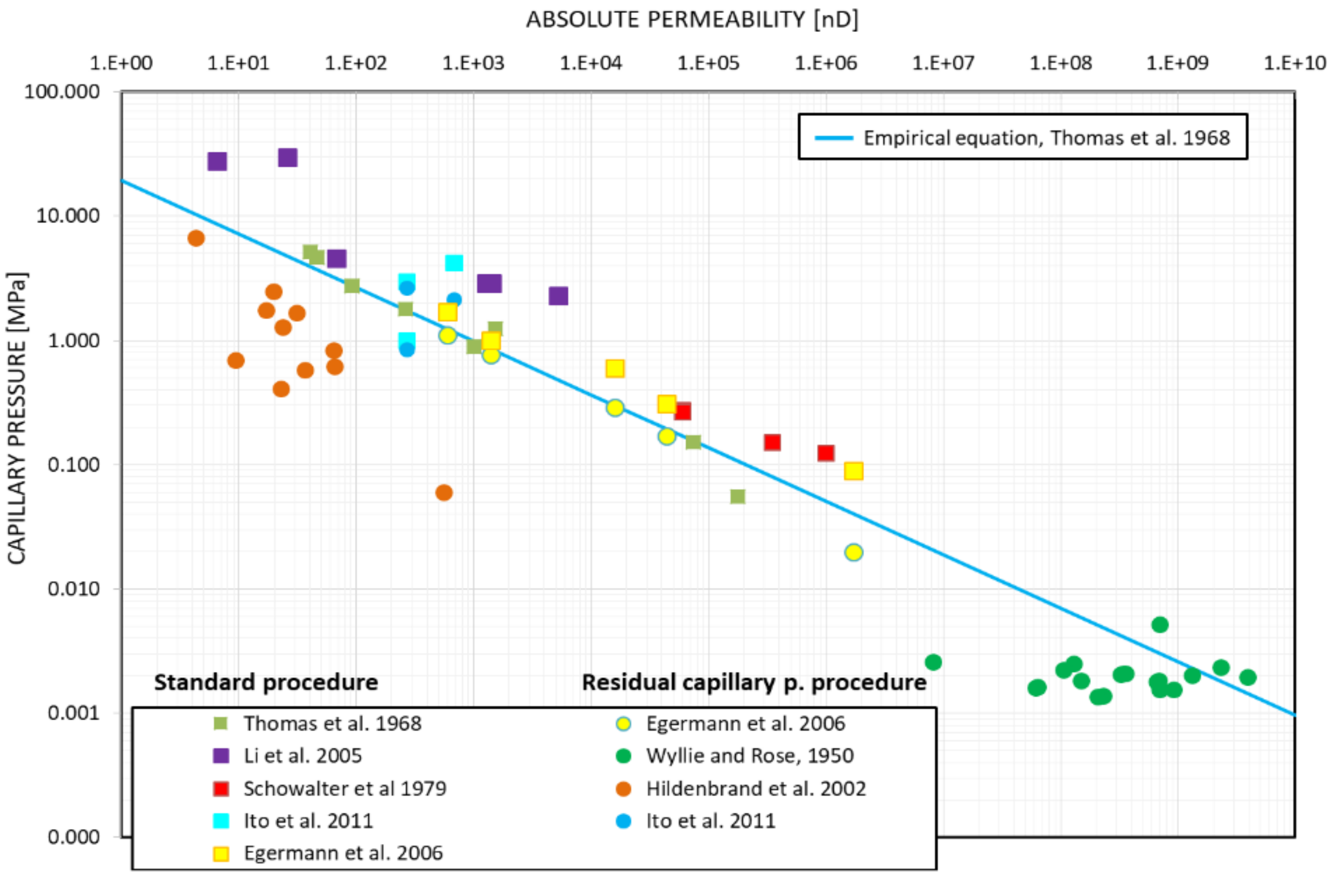

Fig. 6 – Comparison between the results from the standard and from the residual procedures; Thomas et al. (1968) empirical equation.

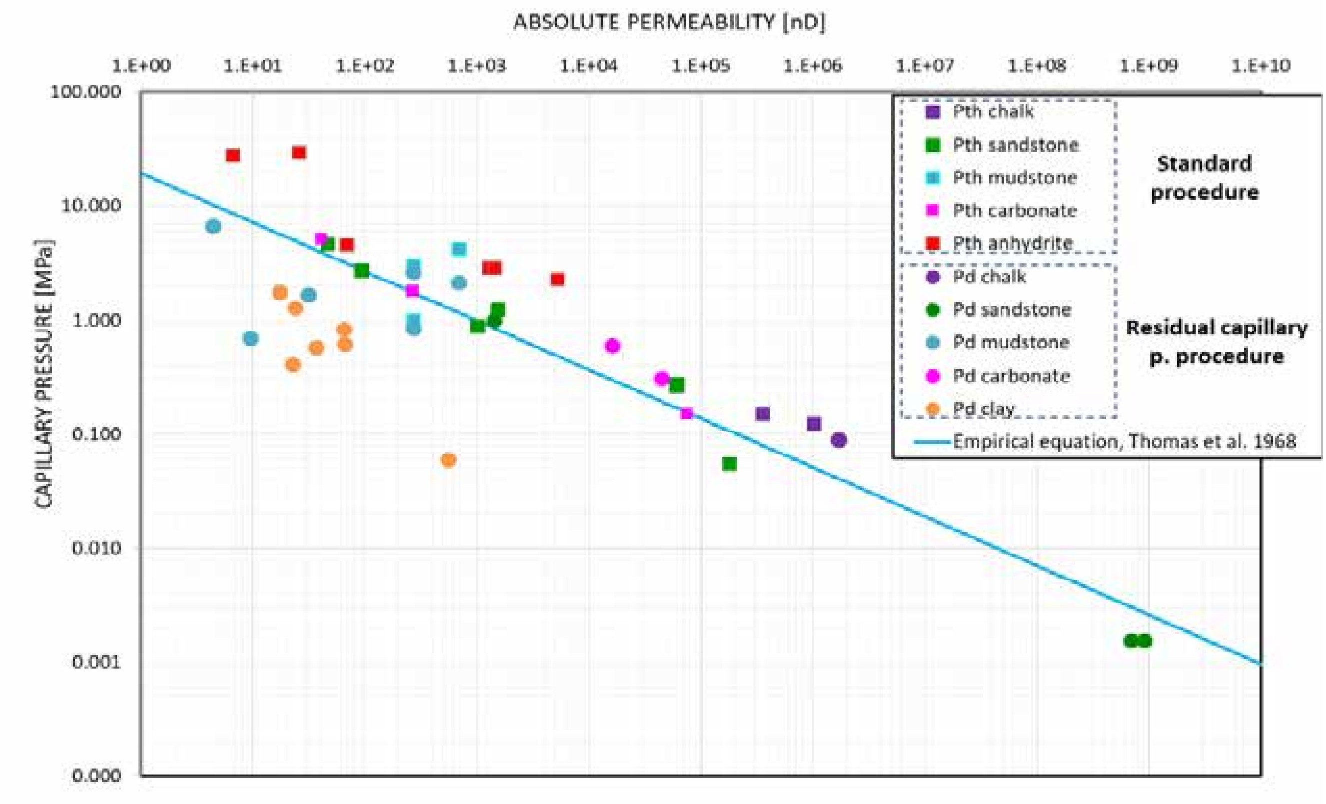

Fig. 7 – Comparison between the results from the standard and from the residual procedures according to the lithological categorization; Thomas *et al.* (1968) empirical equation.

small due to the tightness nature of the investigated materials.
3. Considering the lithological effects, the experimental results on the clay (evaluated only via the residual approach) and the anhydrite (evaluated only via the standard approach) are the most disperse. Each lithology is investigated only by a single publication so no further comparative analyses are possible. Even so, it can be pointed out that all the results from Hildenbrant *et al.* (2002) underestimate the general trend, no matter about the investigated lithology (clay or mudstone).

5. Conclusion

The most accredited and widely adopted experimental procedures for the evaluation of the formation sealing efficiency are the standard or step-by-step approach and the residual capillary pressure approach. They evaluate two differ parameters, which, at least, might coincide: the breakthrough or the threshold pressure, the first, and the minimum displacement pressure or the residual pressure difference the latter. Both parameters are capillary pressure. In order to evaluate the effect of adopted lab procedures, the present paper collects, categorizes and analyzes experimental data from the technical literature, obtained performing standard or residual tests (or both) on different lithologies, in reservoir thermodynamic condition, via N_2-brine fluid system. The results show an acceptable coherency between data from the two procedures within a range of permeability of [10^2-10^5] nD. The highlighted discrepancies seem to be more case sensitive rather than attributed to the adopted procedure or the investigated lithology: crucial aspects, which required more investigation, could be related to the effects of the thermodynamic conditions and of the accuracy and the precision of the measures. Furthermore, the comparison between the experimental data and the empirical correlation defined by Thomas *et al.* (1968) shows a satisfactory agreement, for the permeability range in which the equation was defined. Even if the equation was derived only from standard test measurements on a limited number of lithologies, it turns out to be a reliable predictive approach for the identification, at least, of the order of magnitude of the threshold pressure (or the residual pressure difference).

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