J. Mater. Environ. Sci. 5 (3) (2014) 931-936
ISSN : 2028-2508
CODEN: JMESCN

Relationships between porosity and permeability of calcarenite rocks based on laboratory measurements

Abdelaali Rahmouni[1*], Abderrahim Boulanouar[1], Mohammed Boukalouch[1], Yves Géraud[2], Abderrahim Samaouali[1], Mimoun Harnafi[3], Jamal Sebbani[1]

[1]*Laboratory of Thermodynamics, Faculty of Science, Mohammed V University Agdal, P.O. Box 1014, Rabat, Morocco.*
[2]*University of Lorraine, ENSG, UMR 7359-GeoRessources, Nancy Cedex, France.*
[3]*Department of Earth Science, Scientific Institute, Mohamed V University Agdal, Rabat, Morocco.*

Received 28 Apr 2013, Revised 3 Mar 2014, Accepted 3 Mar 2014
** Corresponding author. E mail: a.rahmouni@yahoo.fr*

Abstract
Fluid transport into porous materials is an area of study relevant to many scientific and engineering fields such as hydrogeology, geoenvironmental engineering, petroleum engineering, chemical engineering and physics. Permeability and porosity are two of the primary properties that control the movement and storage of fluids in rocks. They represent important characteristics of materials. In the present study, basic petrophysical properties of calcarenite rocks were measured, and their relationships are discussed. Permeability and mercury intrusion porosity, and pore size distribution were determined. Furthermore, bulk and particle densities of rocks were determined. Permeability and porosity are closely related to each other in very good direct proportional relationship, i.e., with increasing porosity, permeability increases as well. This relationship is influenced by other rock properties, such as the amount of open and closed pores within the rock sample, as well as pore size, and distribution. From this point of view, it is necessary to study these petrophysical properties of calcarenite rocks that are commonly used for historical monuments, because this enables an overall analysis of rocks and its possible use for engineering constructions and renovation of historical monuments.

Keywords: Calcarenite rocks, Porosity, Permeability, Mercury intrusion porosimetry, Bulk density.

1. Introduction
Permeability is one of the key petrophysical parameters for the management of hydrocarbon and geothermal reservoirs as well as for aquifers. However, its magnitude may vary over several orders of magnitude, even for a single rock type. Moreover, permeability is very sensitive to changes in overburden pressure or to diagenetic alterations. For instance, drastic permeability reductions result from the growth of minute amounts of secondary clay minerals on quartz grains, since this changes the geometry of the hydraulic capillaries. Permeability and porosity are two of the primary properties that control the movement and storage of fluids in rocks. They represent important properties of materials. On the basis of the known permeability and porosity, possible influences of water on an engineering construction are considered. Furthermore, knowledge of permeability and porosity is necessary at water leakages, at the structural foundation in order to evaluate affluent to a foundation pit, and in terms of the design of waterproofing of buildings. Permeability and porosity are also very important indicators for the utilization of various kinds of rocks [1-5].
Permeability of porous media is usually expressed as function of some physical properties of the interconnected pore system such as porosity and tortuosity. Although it is natural to assume that permeability values depend on porosity, it is not simple to determine which is the appropriate relationship since this would require a detailed knowledge of size distribution and spatial arrangement of the pore channels in the porous medium. Permeability depends on the continuity of pore space whereas porosity basically signifies the availability of a pore space, and as such there is no direct relationship between the two. It is also therefore possible to have a very high porosity without having any permeability and similarly a low porosity rock may have a high permeability, as in the case of a micro-fractured carbonate where most of the relatively unimpeded flow takes place through the micro-fracture [6].
Many correlations between permeability, k, and other petrophysical properties have been reported. Purcell [7] presented a method of obtaining mercury injection P_c curves for core samples and derived a relationship between permeability and P_c curves. Certain correlations have been developed between sound wave velocity and the measured porosity and permeability at different cement concentrations, pore size distributions and compaction pressures [8]. Nabawy et al. [9] studied the relationships between helium porosity and the uncorrected air permeability from the

routine core analysis, and the various parameters derived from mercury injection– capillary pressure curves using multiple regressions. Saar [10] compared the permeability of vesicular basalts with the predictions of theoretical models for given porosities and microstructural properties. Gamage et al. [11] developed the permeability–porosity relationships based on sediment type and grain size distribution. Variations observed in permeability – porosity relationships were assessed based on the effect of interlayer water in smectite and the general compaction history. Calcarenite is used mainly for various purposes in the building industry, for the renovation of historical monuments, stonework, sculptures… etc. Limestones are used as facing material and for the restoration of historical buildings. Permeability and porosity have an impact on rock weathering, which affects the field of engineering utilization. Permeability is one of the rock properties that are necessary for considering the solving of hydrological and hydrogeological problems by methods of numerical and physical modelling [12-15].
In this paper, an experimental procedure to measure permeability and porosity on a set of samples from sedimentary rocks is presented. The paper investigates the relationship between permeability, porosity and bulk density for all samples.

2. Materials and methods

The materials used in this study present sample are sedimentary rocks mostly composed of calcarenite, collected from a quarry near the town of Rabat, Morocco. The monuments of Rabat are all constructed by the Pliocene-Quaternary calcarenite that constitutes the basement of the whole region. A few kilometers to the north of the city, quarries still provide this ornamental rock widely used by stone craftsmen. It is characterized by variable and high porosity (18–47%) and thus an elevated permeability [16-19]. Its chemical composition is very rich in calcium carbonates and its rough surface allows a high receptivity to the atmospheric gaseous pollutants and to hydrous marine sprays charged with various salts. Meanwhile, this material is characterized by good mechanical loads (stresses and shocks) [17].
Porosity of porous medium describes the fraction of void space in the rock, where the voids may contain air or water. The porosity is defined as the ratio of the volume of voids expressed as a percentage of the total (bulk) volume of a rock, including the solid and void components. Porosity is calculated from the derived formula [20]:

$$\phi = \left(1 - \frac{\rho_d}{\rho}\right) \times 100 \qquad (1)$$

where ρ_d is bulk density of the dry specimen and ρ is particle density.
Bulk density can be determined from a regular specimen by stereometric method. Particle density, an average mass per unit volume of the solid particles in a rock sample, is usually determined by applying the mercury intrusion [21].
Permeability and porosity depend on pores in the rock. There are two discerned typologies of pores in rocks: closed and open pores. Closed pores are completely isolated from the external surface, not allowing the access of external fluids in either the liquid or gaseous phase. Closed pores influence parameters such as density and mechanical and thermal properties. Open pores are connected to the external surface and are therefore accessible to fluids, depending on the pore characteristics/size and the nature of fluid. Open pores can be further divided into dead-end or interconnected pores. The percentage of interconnected pores within the rock is known as effective porosity. Effective porosity excludes isolated pores and pore volume occupied by water adsorbed on clay minerals or other grains. Total porosity, determined from formula (1), is the total void space in the rock, whether or not it contributes to fluid flow.
One of the most important parameters is the pore size distribution. Pores are classified according to four groups depending on the access size: micropores, with size less than 0.4 µm diameter; mesopores, ranging between 0.4 and 8 µm diameter; macropores, which are in range from 8 µm to 60 µm diameter and megapores in size over 60 µm [9].

2.1. Mercury porosimetry

Method of the high pressure porosimetry is based on phenomenon of the mercury capillary depression, where the wettability angle is > 90° and mercury leaks into pores by the effect of pressure. Mercury volume infiltrated into a porous system is generally interpreted as total pore volume in measured specimen. Relationship between actual pressure P and cylinder pore radius R is expressed by Washburn equation [22]:

$$P = \frac{2\sigma\cos\phi}{R} \qquad (2)$$

where P [Pa] is an actual pressure, R [nm] half-length distance of two opposite walls of a pore expressed by an effective radius, σ surface tension of mercury [480.10^{-3} $N.m^{-1}$] and ϕ contact angle [141.3°].
These porosimetry measurements were performed using an apparatus called Micromeritics Pore Sizer 9320 which makes it possible to inject mercury with pressures ranging between 0.001 and 300 MPa. So the access threshold ranges between 400 and 0.003 µm. This technique determines the connected porous volume and its distribution according to the injection pressure and the thresholds access.
Cylindrical samples, of 25 mm length and 20 mm in diameter, were dried at 60°C for 48 hours, weighed and placed in an injection cell. After a degasification step under a 50 µm mercury depression, the injection cell is filled with mercury, and then the vacuum is broken gradually until atmospheric pressure. The intrusion measurement, i.e., the volume of mercury

injected into the sample, is made for low pressures (between 0.001 and 0.15 MPa) and for high pressures (between 0.15 and 300 MPa). The pressure rises are carried out in stages; after each stage, the injected mercury volume is measured. From these data, it is possible to determine the saturation curve according to the injection pressure.

2.2. Permeability

We used a relatively new surface gas permeameter for making gas permeability measurements on the surface of substrate specimens described above. The permeameter TinyPerm-II, made by New England Research, Inc. It is a unique hand-held device that can characterize the permeability of rocks and soils or the effective hydraulic aperture of fractures in situ on outcrops as well as on laboratory specimens [23]. The apparatus is capable of making permeability measurements ranging from 0.01 to 10 darcies for matrices and 10 μm to 2 mm fracture apertures [23]. Although the measurements could have been made in the field, all measurements reported here were made on the specimens in a laboratory.

This device uses Darcy's law to compute the permeability. Brown and Smith [24] showed that the permeability can be determined using the following relationship:

$$\frac{Q}{P_0} = -\frac{kA}{\mu L} \tag{3}$$

where Q is the net air flow into the piston syringe, P_0 is the applied pressure, which remains constant, A is the inlet area for air flow, μ is the viscosity of the gas (air), and L is the length. Since the values of A, L and μ are known; and Q and P_0 are measured, equation (3) may be solved for permeability, k.

The testing procedure is straightforward; the operator presses a rubber nozzle against the specimen and withdraws air from it with a single stroke of a syringe. As air is pulled from the sample, a microcontroller unit simultaneously monitors the syringe volume and the transient vacuum pulse created at the sample surface. Using signal processing algorithms, the micro-controller computes the response function of the sample/instrument system. Key characteristics of this response are displayed on the liquid crystal display (LCD) screen. Theory shows relationship between the response function and permeability; and either matrix permeability or effective fracture flow aperture may be determined from the calibration charts and tables provided [23, 24].

The response function is related to permeability k:

$$T = -0.8206 \log_{10}(k) + 12.8737 \tag{4}$$

where T is the value of the response function and the recorded output from the mini-permeameter. For each sample point 3–6 measurements were made, in order to ensure the quality of the data. For each sample point, an average of the measurements was calculated and used as the representative value for that point.

Six rock samples were tested, and values of the permeability, particle and bulk densities, and porosity are listed in Table 1.

Table 1: Petrophysical properties of calcarenite rocks.

Samples	Permeability, k (m^2)	Bulk density, ρ_d (g/cm^3)	Particle density, ρ (g/cm^3)	Porosity, ϕ (%)
1	2.28E-11	1.64	2.38	31.07
2	3.34E-11	1.59	2.38	33.50
3	1.74E-11	1.68	2.39	29.82
4	1.40E-11	1.75	2.35	25.69
5	4.37E-11	1.60	2.47	35.07
6	5.04E-11	1.60	2.49	35.83

The values of the presented rock properties were predominantly determined as an arithmetic average of rock specimen tests. For each specimen, the permeability and particle and bulk densities were measured, and porosity calculated. The morphology of the porous medium obtained by mercury porosimetry gives an appearance pore distribution of the material. The samples have sufficient volume to be representative of the material. The results are shown in Figures 1-4 and Table 2.

3. Results and discussion

The results of permeability and porosity of the rocks were analyzed using the least squares regression method. The equation of best fit line and coefficient of determination (R^2) were determined for each regression. The values of permeability of the calcarenite rocks were correlated with the porosity of the rocks.

A very strong correlation between permeability and porosity of the rocks was found. It can be observed that there is linear relation between the permeability of rocks with porosity of the calcarenite rocks. The results of regression equations and the coefficient of determination are presented in Figure 1.

Permeability and porosity are in a close relationship that depends on the amount of void space in the tested material. It is widely accepted that permeability is greatly affected by microstructure, which is, in this context, defined in terms of pore and crack structures. So it could be supposed that with increasing porosity, the permeability should increase as well. But there are some other facts to note when speaking about this relationship. Therefore, permeability of porous material is influenced not only by porosity, but also by shape and arrangement of pores, or by the amount of clayey component [25, 26].

It is necessary to distinguish between total and effective porosity. We are not able to make assumptions on permeability of tested material from values of total porosity, due to the fact that it is the total void space in the rock. A rock may be highly porous, but if the voids are not interconnected, fluids within the closed (isolated) pores cannot leak, e.g. shales and pumice rock, both are highly porous but impermeable.

The pore size distribution is very important petrophysical property controlling the fluid flow. To clarify the relationship between permeability and porosity; pore size and pore size distribution were determined for selected rock samples. Pore dimensions cover a very wide range. Within our research, two samples of calcarenite, which have approximately the same order values of permeability, but different porosities, were tested by mercury porosimetry for pore size distribution.

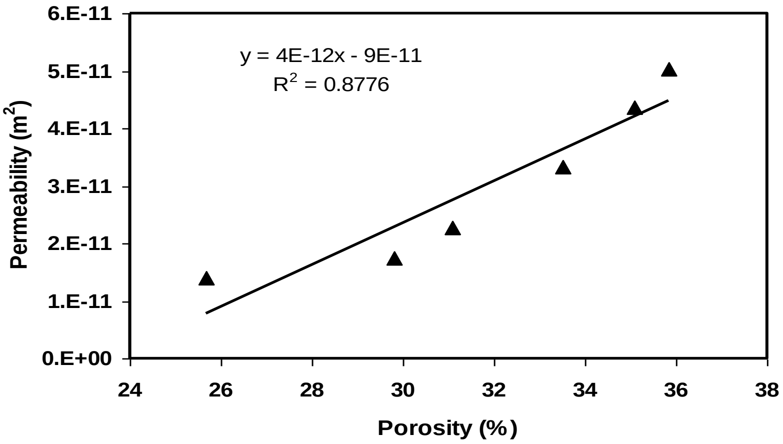

Figure 1: Permeability versus porosity of calcarenite rocks.

Figures 2 and 3 show the polymodal nature of the pore size distribution of the studied stones. The relevant features here are the number and kind of pore families, rather than pore volume frequency.

As we can see from Figures 2 and 3, sample 2 has a more uniform distribution in the range of pore size. The megapores of this sample should not exceed 65.06 % in total. It has a very broad spectrum of porosity with significant mesoporosity. The prevailing part of pores belongs to the megapores, which can create main transport ways for liquid. In the case of sample 3, the distribution of pore size is different. The megapores of this sample about 58 % in total. The dominant part of pores belongs to the mesoporosity, and microporosity, which is also very important.

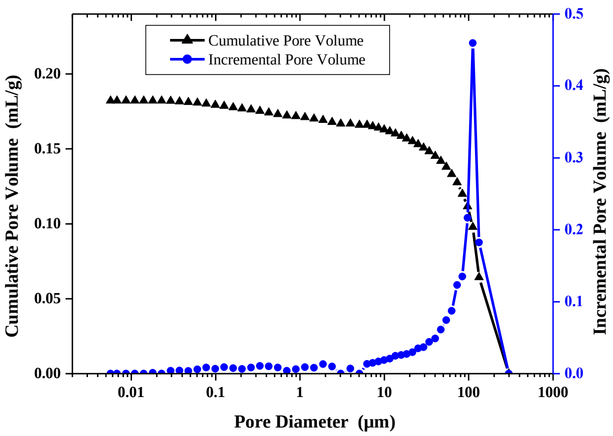

Figure 2: Pore size distribution of sample 2.

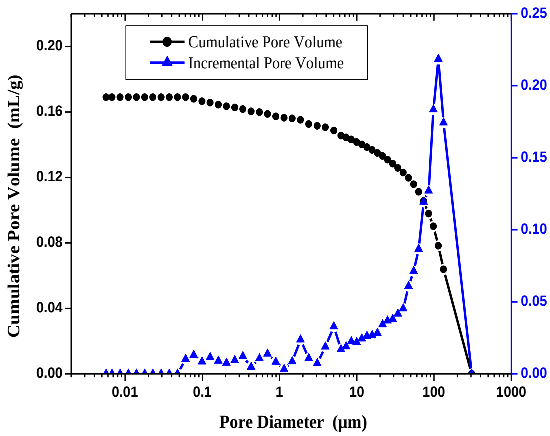

Figure 3: Pore size distribution of sample 3.

The distribution of pore volumes of sample 2 is unimodal (a single dominant family of pores resulting in a single point of inflection in the curve of injection), it is possible to determine a radius access or threshold pore: it corresponds to the radius access of pore that to a low pressure increment provides access to a large pore volume. It is graphically determined on the first injection curve as the radius corresponding to the inflection point of the curve or by the method of tangents [21, 22]. The threshold pore can also be viewed on the curves representing the increment of mercury injected for each access radius. The injection curves with multiple turning points illustrate multimodal porous networks where several families ray access to the pores coexists.
Average pore diameter is usually used as a representative parameter of the pore size distribution. In case of sample 2, average pore size diameter is 1.95 µm; for sample 3, average pore size diameter is 2.26 µm (Table 2).
It is well known that mineral admixtures affect permeability. The basis for this effect can be understood in terms of the formation of a large amount of porosity in the mesopore range.

Table 2: Petrophysical properties of two calcarenite samples.

Samples	Permeability k (m^2)	Bulk density ρ_d (g/cm^3)	Particle density ρ (g/cm^3)	Porosity ϕ (%)	Average pore size diameter (µm)
2	3.34E-11	1.59	2.38	33.50	1.95
3	1.74E-11	1.68	2.39	29.82	2.26

Finally, there are some other characteristics of samples we observed, such as the relationship between bulk density and porosity. We have obtained the generally known fact from calcarenite rock samples. The relationship between bulk density and porosity of all 6 tested samples can be seen in Figure 4. We can clearly identify that with decreasing bulk density, the porosity of the sample increases. It is due to the small differences in particle densities, which are not dependant on porosity, but only on modal composition. The modal compositions of calcarenite samples are assumed to be approximately the same.

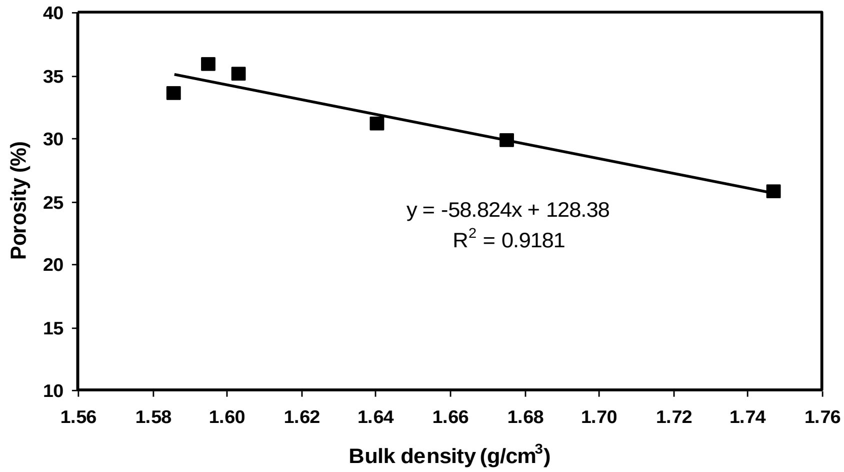

Figure 4: Bulk density versus porosity of calcarenite rocks.

The relationship between bulk density and porosity of all tested samples is shown in Figure 4. Calcarenite has high porosity. The permeability of the rocks above depends on the structured nature of minerals forming the solid matrix. The permeability of these rocks is in the range of 10^{-11} m^2.

Conclusions

Laboratory measurements of the petrophysical properties of calcarenite rocks were carried out and their relationships were discussed. The above-mentioned petrophysical properties of rocks, such as permeability, porosity, pore size distributions and bulk density are very important characteristics. The values of permeability of the calcarenite rocks were correlated with the porosity of rocks.
Permeability of rocks is determined by microstructure, which is, in this context, defined in terms of pore and crack structures. Permeability of porous media is usually expressed as function of some petrophysical properties of the interconnected pore system such as porosity, bulk and particle density. Although it is natural to assume that permeability values depend on porosity, it is not simple to determine which is the appropriate relationship since this would require a detailed knowledge of size distribution and spatial arrangement of the pore channels in the porous medium.

Acknowledgements-The authors would like to thank Dr. Amin Esmail Khalili, School of Physics, USM Malaysia, and Dr. Bassem S. Nabawy, Department of Geophysical Sciences, National Research Centre, Cairo, Egypt, for their many incisive comments that improved significantly this work.

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