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Deriving the Pore Structure of Selected Jordanian Building Limestones

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Abstract

Making water absorption as the independent variable in the equations of Moh'd and Cranganu (2012), we have used these equations to predict the pore structure (porosity, pore size distribution, seepage radius, specific surface area, tortuosity and permeability) of 12 clean Jordanian building limestones. In the present work, first, the stones were subdivided into unimodal and bimodal pore networks (by plotting their porosity against the bulk volume water or modified saturation). Then, the 24-hour water absorption was used to derive the pore structure of unimodal and bimodal porosity limestone. The two chalky samples (Izrit and Hatem) stones gave anomalous results. This can be explained by the fact that the original Russian database, which the present work is based upon, did not include any chalky samples. The results are not thus recommended to be extended to the to chalky limestones; it is however recommended that a similar research be carried out employing a representative number of chalk samples for deriving the pore structure of chalk from water absorption.

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Keywords: pore structure, tortuosity, surface area, permeability, water absorption, Jordanian building limestone.

1. Introduction

Although it is known that water behavior in limestone is a function of limestone's pore structure which is largely controlled by the texture of the rock and also despite the fact that water absorption (amount and/or rate) measurement is required for most building material (stone, aggregate and concrete) standards, very little work has been done to quantify the water absorption-pore structure relationship (Coskun and Wardlaw, 1995; Winslow, 1987).

Pore structure characterization (amount, size, shape, connectivity and distribution) characterization in carbonaterocks is important in many applications related to oil industry (Dullien, 1979; Asquith, 1985; Jordy, 1992; Chillingarian et al., 1992) and to ground water aquifer exploitation and pollution (Cander, 1995). As many industrial applications of limestone are controlled by purity, grain size and internal surface area they may directly or indirectly be influenced by pore structure (Hartman and Coughlin, 1974). In limestone and dolomite, metallic ore deposits are usually present in both primary and secondary pore types (Anderson and Macqueen, 1982).

In civil engineering, suitability of carbonate rocks as building materials (building stone, aggregate and concrete and asphalt mixes) and their frost- and salt-durability are largely controlled by their pore structure (Leary, 1983; Cnudde et al., 2009). Knowledge of pore structure is critical for the success of restoration of historic buildings and archeological sites (Ashurst and Dimes, 1990).

It is well-known that the amount of water absorption depends on porosity and the presence of absorptive mineral

species such as clay minerals.

Moh'd and Cranganu (2012) investigated the water absorption in limestone as a new tool for pore space characterization. They used a small Russian database of clean limestone and dolostone to relate, using cross-plotting and multiple regression analysis methods, the rate of water absorption to pore structure parameters in unimodal and bimodal pore networks. Relationships have been established between porosity and pore size distribution, seepage radius, specific surface area, tortuosity and permeability with water absorption. These researchers found that most of the petrophysical properties can be easily derived from the amount of 24 hour water absorption. However, porosity and bulk volume water or modified saturation (= porosity * saturation) need also to be known or estimated so as to define the uni- or bi-modality of the pore space. Thus, a new and simple method is proposed to derive approximate values of most petrophysical properties using weight difference after submerging a carbonate sample in water for 24 hours. Porosity can be measured or derived from density. Pore network unior bimodality was found to have a stronger effect on the petrophysical properties than carbonate lithology type.

In clean carbonates, either capillary imbibition or total immersion causes the water sorption of porous material under natural conditions. The water sorption from total immersion can be considered as a generalized capillary sorption of all rock material surfaces (Bellanger et al., 1993).

The ratio between the natural capacity of a rock to absorb water and its absolute porosity is termed as saturation coefficient (Hirschwald, 1912); this principle has been

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introduced in connection with frost damage caused by the 10% expansion that occurs on freezing.

In the present work, it is intended to derive the pore structure of selected Jordanian building limestones using database of Moh'd and Cranganu and the 24-hour water absorption as an independent variable after dividing their pore structure into unimodal or bimodal porosity types. Pore structure elements that are derived include: pore size distribution, seepage radius, specific surface area, permeability, and tortuosity. 12 Jordanian building limestones are studied including: Izrit, Hatem, Ishtafina, Yanabi, Hallabat, Tafih, Sahrawi, Mafraq, Jazeira, Sat'h, and Siwaqa and Mujeb travertine.

2. Methods and Materials

Water absorption, porosity and water saturation of the studied samples were measured at the laboratories of the QMW (University of London) and Building Research Establishment (BRE) in the United Kingdom (Moh'd, 1996). They were also employed in other publications of the author (Moh'd, 2002; 2007; 2008).

To subdivide the suite of the studied samples into those of unimodal and bimodal pore networks, porosity was plotted against modified saturation (Bellanger et al., 1993) (Figure 1). The following properties were multiplied by porosity to get an idea about the bulk volume water after a 24-hour saturation (modified 24 hour saturation), and pore size percentage less than 0.2 μ m (modified less) and more than 0.2 μ m (modified more). According to Moh'd (1996), most saturation takes place during the first hour of soaking. Thus, the 24-hour absorption was considered here to be roughly equivalent to saturation after 16 hours (used in the Russian database).

The following definitions of terms are used:

- Water absorption %: the weight of water absorbed by the rock after 24 hours of immersion in water divided by its oven-dried weight expressed as a percentage of its oven-dried weight.
- Modified water absorption: water absorption multiplied by porosity.
- Porosity%: the percentage of volume of voids over the total volume of rock.
- Saturation %: the percentage of pore volume, which can be filled with water after immersion in water for 24 hours.
- Modified saturation (msat): is saturation multiplied by porosity.
- Effective porosity % (effecpor): indicates interconnected pores and is the product of water absorption and dry density.

- More than 0.0002 mm pore diameters (more): percentage of this pore size portion.
- Modified more than 0.0002 mm pore diameters (mm): percentage of this pore size portion multiplied by porosity.
- Less than 0.0002 mm pore diameters (less): percentage of this pore size portion
- Modified less than 0.0002 mm pore diameters (ml): percentage of this pore size portion multiplied by porosity.
- Tortuosity (tort): the ratio of total path covered by an electric current flowing in the pore channels between two electrodes to the straight-line distance between the electrodes.
- Permeability (perm): in millidarcies, the ease with which fluids pass through the rock.
- Specific surface area (ssa): surface attributed to tortuosity (m²/g).
- Seepage radius (seep rad): the radius of pore channel in µm.

The following equations have been derived from the original Russian database to derive the pore structure of Jordanian limestones:

For limestones with unimodal porosity:

Modified more = $0.3357*$ modified saturation ^{1.4453}	$r^2 = 0.87$
Modified less = $0.0497*$ modified saturation	$r^2 = 0.95$
+ 0.9588	
Tortuosity = 9.6416 / modified saturation ^{0.3902}	$r^2 = 0.90$
Permeability = 6 E-7* modified saturation ^{5.5998}	$r^2 = 0.90$
Specific surface area = 0.59938 Ln modified	$r^2 = 0.86$
saturation + 18.243	
Seepage radius = $0.2156 \text{ e}^{0.2958 \text{ modified saturation}}$	$r^2 = 0.93$
For limestones with bimodal porosity:	
Modified more = 2.293* modified saturation +	$r^2 = 0.97$
0.1807	
Modified less = - 0.1292 modified saturation +	$r^2 = 0.99$
1.5451	
Tortuosity = -0.664 modified saturation +	$r^2 = 0.68$
12.506	
Permeability = $0.0001 \text{ modified saturation}^{4.4167}$	
Specific surface area = -2.3865 Ln modified	$r^2 = 0.80$
saturation + 6.8761	
Seepage radius = 0.7217 modified	$r^2 = 0.93$
saturation ^{1.6747}	

3. Results and Discussion

Using the above-mentioned equations, the results of the pore structure elements calculations are summarized in Tables 1 and 2 along with correlation matrices between the different variables in both the uni- and bimodal porosity types.

Porosity Saturation msat effecpor more mm less ml Seep r tort perm ssa 1.92 0.33 Ballas 9 0.75 1.44 1.46 0.296 0.5687 0.536 1.03 8.36 4.62E-06 18.02 Yanabi 10 1.52 0.80 1.216 1.214 0.293 0.4453 0.671 1.02 8.93 1.79E-06 18.13 0.31 Karak 9 1.79 0.75 1.34 1.224 0.286 0.5124 0.575 1.03 8.60 3.09E-06 18.07 0.32 4.459 2.7825 0.203 0.77 Hayyan 10 5.76 0.75 4.32 0.483 1.17 5.45 2.17E-03 17.37 27.72 Izrit 9 0.84 23.29 23.56 1.146 31.766 0.076 2.12 2.82 27.18 16.35 211 Hatem 10 20.87 0.78 16.28 16.02 0.907 18.9302 0.085 1.77 3.25 3.66 16.57 27

Table 1. Derived pore structure elements and correlation matrix in rocks with unimodal pores

Table 1 continued

	Porosity	Saturation	msat	effecpor	more	mm	less	ml	tort	perm	ssa	seep rad
Porosity	1.00											
Saturation	0.72	1.00										
msat	1.00	0.74	1.00									
effecpor	1.00	0.75	1.00	1.00								
more	1.00	0.71	1.00	1.00	1.00							
mm	0.99	0.78	1.00	1.00	0.99	1.00						
less	-0.86	-0.37	-0.85	-0.85	-0.89	-0.81	1.00					
ml	1.00	0.74	1.00	1.00	1.00	1.00	-0.85	1.00				
tort	-0.94	-0.52	-0.93	-0.93	-0.95	-0.89	0.98	-0.93	1.00			
perm	0.84	0.84	0.86	0.87	0.84	0.90	-0.60	0.87	-0.69	1.00		
ssa	-0.97	-0.59	-0.96	-0.96	-0.98	-0.94	0.96	-0.96	0.99	-0.74	1.00	
seep rad	0.84	0.84	0.86	0.87	0.84	0.90	-0.59	0.86	-0.68	1.00	-0.74	1.00

Table 2. Derived pore structure elements and correlation matrix in rocks with bimodal pores.

	Porosity	Saturation	msat	effecpor	more	mm	less	ml	tort	perm	ssa	Seep r
Hallabat 10	14.32	0.59	8.449	9.279	1.365	19.55	0.031	0.45	6.89	1.24	1.78	25.73
Tafih 10	17.2	0.56	9.632	9.59	1.295	22.27	0.018	0.31	6.11	2.22	1.47	32.05
Travert 2	1.34	0.54	0.723	0.563	1.373	1.84	1.082	1.45	12.02	2.39E-05	7.65	0.42
Ma'an a 10	4.66	0.49	2.283	1.57	1.163	5.42	0.268	1.25	10.99	3.83E-03	4.91	2.88
Ma'an b 9	3.74	0.35	1.309	1.61	0.850	3.18	0.369	1.38	11.64	3.28E-04	6.23	1.13
Sahrawi 10	13.72	0.60	8.232	7.68	1.388	19.04	0.035	0.48	7.04	1.11	1.85	1.52

Table 2 continued

	Porosity	Saturation	msat	effecpor	more	mm	less	ml	tort	perm	ssa	seep rad
Porosity	1.00											
Saturation	0.63	1.00										
msat	1.00	0.69	1.00									
effecpor	0.99	0.66	0.99	1.00								
more	0.46	0.97	0.53	0.50	1.00							
mm	1.00	0.69	1.00	0.99	0.53	1.00						
less	-0.82	-0.24	-0.79	-0.78	-0.02	-0.79	1.00					
ml	-1.00	-0.70	-1.00	-0.99	-0.53	-1.00	0.79	1.00				
tort	-1.00	-0.69	-1.00	-0.99	-0.53	-1.00	0.79	1.00	1.00			
perm	0.96	0.60	0.94	0.94	0.47	0.94	-0.67	-0.94	-0.94	1.00		
ssa	-0.98	-0.62	-0.98	-0.96	-0.43	-0.98	0.90	0.98	0.98	-0.88	1.00	
seep rad	0.78	0.43	0.76	0.80	0.31	0.76	-0.55	-0.76	-0.76	0.86	-0.71	1.00



Figure 1. Unimodal porosity (blue points) fit with a straight line below which (pink points) bimodal porosity lies

Very few studies have been done previously on Jordanian stones apart from the work published by the present author (Moh'd, 2002; 2007; 2008) and by Ahmad (2011). As it can be seen from the very high correlation coefficients in both the uni- and bimodal porosity building stones of Jordan, most pore structure elements can be derived from porosity.



Figure 2. Porosity versus modified more in unimodal pore networks



Figure 3. Porosity versus modified less unimodal pore networks







Figure 5. Porosity versus permeability unimodal pore networks







Figure 7. Porosity versus modified more in bimodal pore networks



Figure 8. Porosity versus modified less in bimodal pore networks



Figure 9. Porosity versus tortuosity in bimodal pore networks



Figure 10. Porosity versus permeability in bimodal pore networks



Figure 11. Porosity versus specific surface area in bimodal pore networks



Figure 12. Porosity versus seepage radius in bimodal pore networks; one bad point was ignored

The two chalky samples (Izrit and Hatem stones) gave anomalous results of seepage radius. This can be explained by the fact that the original Russian database, which the present work is based upon, did not include any chalky samples. Using mercury intrusion porosimetry, Price et al. (1976) measured median pore diameters of the UK chalks ranging from 0.39 to 0.65 μ m and 0.22 μ m in marly chalk. Similarly, Bellanger et al. (1993) reported 0.4 μ m for pore throats of French Jurassic chalk.

3. Conclusions and Recommendations

Most of the elements of pore structure of Jordanian building limestones were derived from the 24-hour water absorption values using 12 equations derived from the original Russian database. Modified saturation is the independent variable in these equations with very high correlation coefficients in both rocks with unimodal pore networks and those with bimodal porous networks. The difference between the present work and that carried by the present author in 2012 was that in the equations of the later work the 24-hour absorption was considered as the dependant variable.

Most pore structure elements can be derived directly from porosity (Figures 1-12). The two chalky samples (Izrit and Hatem stones) with unimodal porosity gave anomalous results concerning their seepage radii. This can be explained by the fact that the original Russian database, which the present work is based upon, did not include any chalky samples. It is thus recommended that the results not be extended to chalky limestones and to carry out a similar research employing a representative number of chalk samples to derive the pore structure of chalk from water absorption.

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