



# **Effect of acidic and basic water on the geomechanical properties of limestone used in decoration of buildings in Iraq**

*Heyam Saleh Daoud\**, *Kamal Ahmad Rashed\*\** & *Farouk Abdullah Rasheed\*\*\**

\* *Irrigation Department, University of Sulaimani, Kurdistan Region-Iraq, [hyam.daoud@univsul.edu.iq](mailto:hyam.daoud@univsul.edu.iq)*

\*\* *Civil Department, University of Sulaimani, Kurdistan Region-Iraq, [kamal.rashed@univsul.edu.iq](mailto:kamal.rashed@univsul.edu.iq)*

\*\*\* *Irrigation Department, University of Sulaimani, Kurdistan Region-Iraq, [farouk.rasheed@univsul.edu.iq](mailto:farouk.rasheed@univsul.edu.iq)*

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## **Article info**

Original: 28.07.2015  
Revised: 06.11.2015  
Accepted: 12.11.2015  
Published online:  
20.06.2016

### **Key Words:**

limestone, physical-  
mechanical  
properties, pH,  
atmospheric  
pollution.

## **Abstract**

The physico-mechanical properties of rock play an important role in planning and designing of civil constructional work. A type of stone used in decoration of buildings in Iraq traditionally known as (Halan rock), has been taken to check variation in its physico-mechanical properties. Thirty two specimens of dimension 4.8cmx4.8cmx10cm of fossiliferous limestone of Lower Fars Formation were collected. Different pH solution from (pH1, pH3, pH5, pH7, pH9, pH11 and pH13) have been used to find the durability of the stone. Laboratory test also includes effective porosity, both dry and saturated unit weight, water absorption, uniaxial compressive strength, point load strength, and modulus of elasticity together with thin section analysis. The results indicated that the limestone appears to be less resistant to the atmospheric factors. The uniaxial compressive strength and point Load strength decrease under acidic and alkaline conditions.

## **Introduction**

Building stones are used as an important material in the lives of human beings and history of civilization. They are also used in both the construction of modern buildings and in conservation as a replacement material for the reconstruction of monuments [1, 2]. Stone monuments represent an important part of global cultural heritage. The awareness of people is increased for weathering damage to historic monuments and acid rain pollution. Therefore, their conservation is receiving growing attention and has become a focus of research in many fields [3, 4]. The natural stones which are being used in interior and exterior of buildings are the important materials in the world. They should have a good quality related to their physico-mechanical properties for planning and designing of construction works in modern structures and buildings [5, 6]. Carbonate rocks are a class of sedimentary rocks, composed primarily of carbonate minerals. The two major types are limestone, which is composed of calcite or aragonite (different crystal forms of  $\text{CaCO}_3$ ) and dolostone, which is composed of the mineral dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ). Carbonate rocks have been widely used as building

stones in constructions and other purposes since ancient times, e.g. in Rome and across the whole Mediterranean [7]. Carbonate rocks are being used in decoration of buildings. They are one of the most common construction materials in the world [6]. Carbonates rocks are formed in different depositional environments and consist of various amounts of cement, calcium carbonate of biogenic origin and nonbiogenic materials. Thus, the different physical and mechanical properties determine the resistance of these rocks to physical and chemical weathering. The chemical weathering is important due to continuous atmospheric pollution by acidic agents [8, 9]. The common constituents in atmospheric rain are sulfuric acid ( $H_2SO_4$ ), hydrochloric acid (HCl), and nitric acid ( $HNO_3$ ).

The effect of acid solutions on the damage and weathering of carbonate stone has long been recognized [11, 12, 13, 14]. The acceleration in damage of building stones, is due to the atmosphere pollution [15, 16, 17], and climatic factors [18]. The rate of damage and deterioration of building stones exposed to the atmosphere increases in the presence of urban and industrial pollutants [17]. The use of fossil fuels in recent decades, generate gases which contribute to air pollution and during acid rain. Different types of dilute acid are produced depending on the type of gases in the atmosphere especially composed of  $H_2SO_4$ ,  $HNO_3$  and HCl [19]. All carbonate rocks are sensitive to acidic solutions. In acid rain the primary contributions of hydrogen ion besides the natural sources of acidity are sulfurous, sulfuric, and nitric acids, which lower the pH of rain and accelerate weathering processes [20]. The studies of the effect of water on the physico-mechanical behaviour of rock, results revealed that the acidic water changed the behaviour and strength of the rock. The strength decreased linearly from neutral pH to the acidic condition [21].

The purpose of present study is to determine the resistance of limestone from Lower Fars to the dissolution in acidic solutions with concentrations resembling those from the atmospheric precipitants. This limestone is used in Iraq as an ornamental and decorative stone due to its attractive patterns and designs. The weathering may have a degrading effect on the composition soundness and appearance of this stone. The agents of weathering include rain, snow, wind, temperature and atmospheric pollutants. These agents act in combination with one another or with other agents of deterioration [10]. In this study, an attempt has been made to determine the effect of the acidic and alkaline solutions of pH values (pH1, pH3, pH5, pH7, pH9, pH11 and pH13) on the physico-mechanical properties of Lower Fars limestone. The Lower Fars Formation occurs in Iraq, Iran and Syria and it was defined from the Fars Province in Iran and introduced to Iraq by Busk and Mayo in 1918 [22]. The type section has been determined in the Al Fatha Gorge, 10 km north of Baiji town where the Tigris River crosses the Makhul-Hemrin range and the name of the formation has been changed to the Fatha Formation [23]. The thickness of the formation is variable, with a maximum thickness of 600 m in the central part of the basin in Iraq. The formation is an evaporitic sequence and consists mainly of repeated cycles of mudrock, marl, limestone and gypsum, locally with halite. In Iraq, the formation is of Middle Miocene age [22]. The formation contains reserves of limestone and gypsum which is used for construction purposes, especially buildings decoration in Iraq.

## **Materials and methods**

The representative limestone blocks were collected from one of the decoration companies in Sulaimani City. These blocks were cut and prepared in the form of prism 4.8 cm x 4.8 cm x 10 cm in a marble factory. The prepared specimens were put into water for 24 hours until they became saturated, then been weighing. The saturated samples were subsequently placed in an oven for 24 hours at 105C° to eliminate the moisture present within them for using in determination of moisture content, dry unit weight ( $\gamma_d$ ), saturated unit weight and porosity ( $n$ ).

The porosity was calculated from the formula:  $n=1 - (\gamma_d/G_s.\gamma_w)$ , where the specific gravity ( $G_s$ ) was determined in the laboratory. Then the samples were treated and tested under the different pH solutions (pH1, pH3, pH5, pH7, pH9, pH11 and pH13) for 24 hours, to understand the behaviour of limestone used in building decoration in various acidic and alkaline conditions (Fig. 1). The different physio-mechanical properties of limestone of Lower Fars Formation such as, uniaxial compressive strength (UCS), point load strength index ( $I_s$ ), density, and Modulus of Elasticity (Young's modulus), were determined at different pH values in the laboratory.



Fig. 1 The specimens, after taking them out of the solution.

## Discussion and Interpretation

### 1-Petrographic analysis

The microfacies associations identified in thin sections consist of packstone and wackestone (Figs. 2 and 3). The packstone consists of foraminifera, gastropods, bioclasts and green algae. The wackestone consists of benthic foraminifera and bioclasts of gastropods. The composition of limestone according to laboratory analysis, consists of 97% calcium carbonate.

The porosity in carbonate rocks, especially in limestone can be of two types [24]. The first one is the primary porosity, this type can be present between particles or within them and as shelter pores; and the second one is the secondary porosity which is formed as a result of post-depositional dissolution such as vugs porosity or fracturing (Fig. 4). Thus, most of the porosity in limestone is of second type porosity. However, primary porosity may be preserved due to the influx of hydrocarbons into pores [25]. Also, secondary porosity in limestone can be formed at, or near the Earth's surface by freshwater dissolution or in deep environments by chemically aggressive fluids.

In this study, the evaluation of the effective porosity has been identified in two ways: The first one by put the samples in water for 24 hours to reach the saturation conditions and by using the equation  $n=1 - (\gamma_d/Gs.\gamma_w)$ , the porosity was been calculated. In this way, the porosity ranges between 12.61% to 18.28% (Table. 1). The second one is based on the study in thin sections under the microscope, by using the point count method. The samples are impregnated under a vacuum with special blue-coloured resin of low-viscosity, to facilitate observation of porosity once thin sections have been completed.

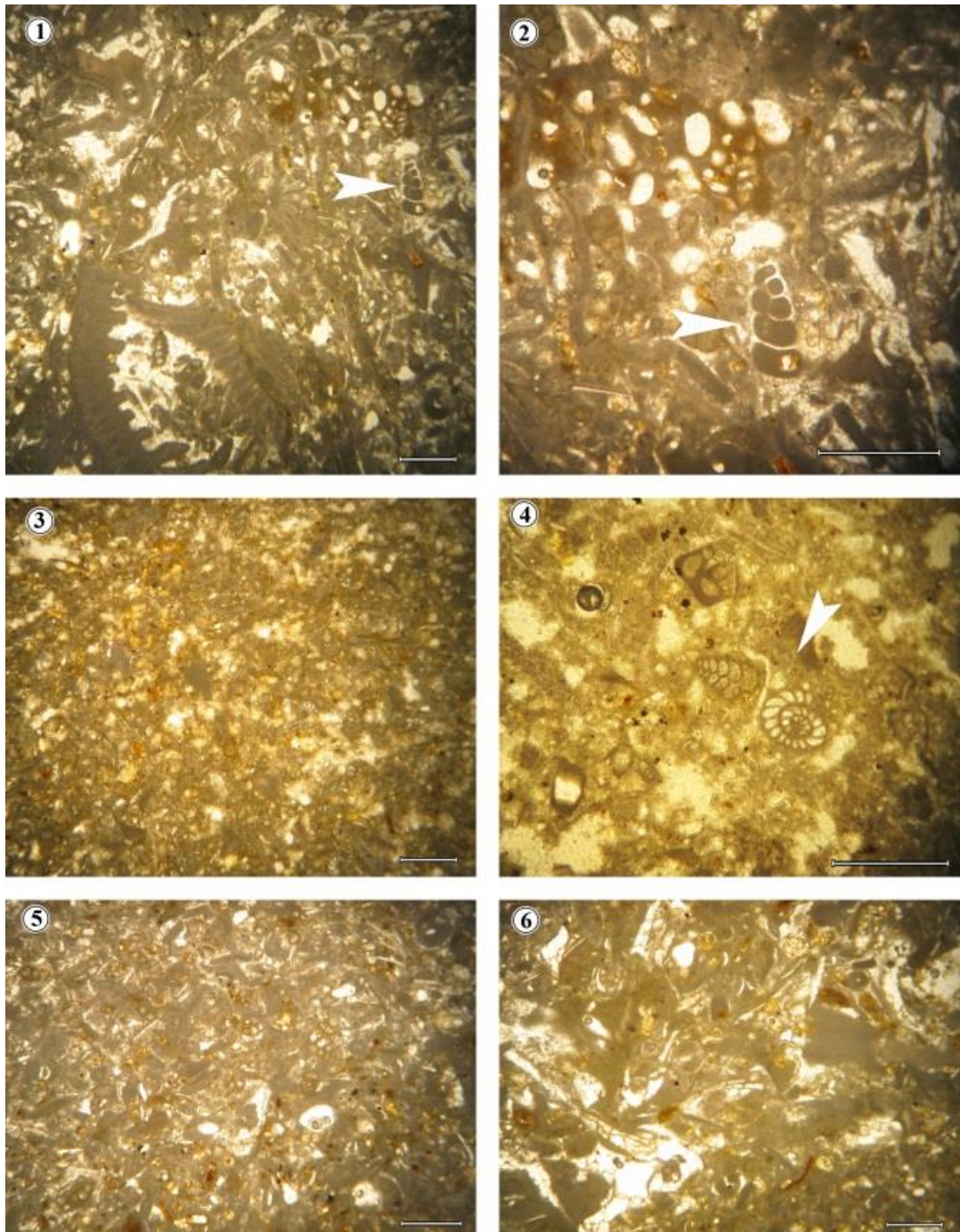


Fig. 2 Photos 1-2 Packstone with foraminifers, gastropods and green algae. Arrows indicate the gastropods. Photos 3-4 Wackestone, the arrow indicates the benthic foraminifera. Photos 5-6 Bioclastic packstone. (bar scale 1mm).

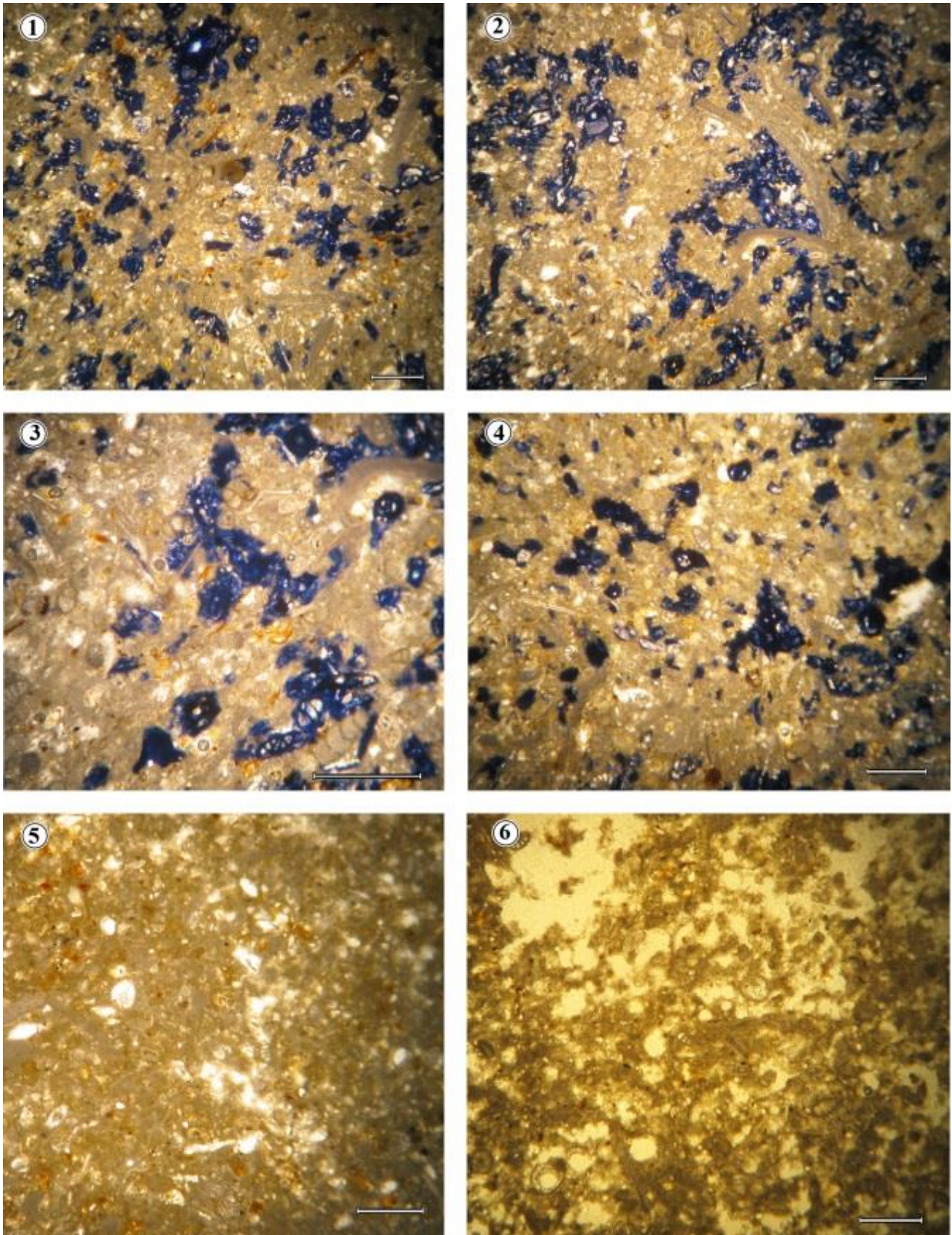


Fig. 3 Effective porosity (blue color). Photos 1-2 porosity 19%, photo 3 porosity 11% and photo 4 porosity 13%. Photos 5-6 Wackestone. Photo 6, the white color due to the epoxy resin that occupies the voids indicates the porosity that reaches 20%. (bar scale 1mm).


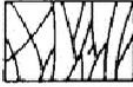
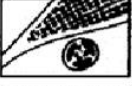


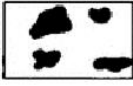


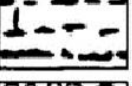


Fabric Selective Porosity		Nonfabric Selective Porosity	
Geometry	Genesis	Geometry	Genesis
 Interparticle	Diagenetic	 Fracture	Deformation
 Intraparticle	Depositional	 Channel	Dissolution
 Intercrystalline	Diagenetic	 Vug	Dissolution
 Moldic	Diagenetic	 Cavern	Dissolution
 Fenestral	Depositional		
 Shelter	Depositional		
 Growth or Framework	Depositional		

Fig. 4 Fabric and nonfabric pore geometries and processes that create them. Modified from Choquette and Pray (1970).

The open pores have been fully filled with blue dye. The vug-type porosity (Fig. 3, photos 1-4 and 6) is more frequently observed and noticed, ranges between 11% to 19%. This porosity type is not related to the fabric, being the result of dissolution due to meteoric waters of grains or intergrain cement during the early, or late diagenetic stages; the result are mm- to cm-sized extremely irregular voids with no definite shape and are caused partly by dissolution-enlargement of interparticle or moldic pores.

## 2- Uniaxial compressive strength test

The samples were tested at different pH values pH1, pH3, pH5, pH7, pH9, pH11 and pH13 to determine the uniaxial compressive strength. The maximum value of UCS (8.43 MPa) was found at pH 7 whereas the lowest values (4.06 MPa, 5.62 MPa) were at 1.0 and 13 pH (Table. 2 and Fig. 5). The uniaxial compressive strength reduction under acidic and alkaline conditions is due to the chemical reaction of the pH solution on the rock composition. It must be mentioned that the value of UCS in the case of the dry state is 20.60 MPa.

In many studies, it was reported that the mechanical strength of rocks decreased under saturated conditions. Reference [26] studied the effect of liquids on mechanical strength and abrasiveness of rocks at different pH values and stated that the mechanical strength of the rock samples decreased when saturated with different liquids compared with their dry values.

Table. 1 Physical properties of Lower Fars fossiliferous limestone.

Sample No.	Saturated Weight (g)	Dry Weight (g)	Moisture Content%	Saturated Unit Weight (g/cm <sup>3</sup> )	Dry Unit Weight (g/cm <sup>3</sup> )	Effective Porosity%
1	521.34	480.95	8.40	2.26	2.09	15.49
2	529.68	485.09	9.19	2.30	2.11	14.76
3	508.64	465.04	9.38	2.21	2.02	18.28
4	532.31	487.97	9.09	2.31	2.12	14.25
5	521.17	478.59	8.90	2.26	2.08	15.90
6	521.23	479.53	8.70	2.26	2.08	15.74
7	518.48	478.28	8.41	2.25	2.08	15.96
8	518.21	478.07	8.40	2.25	2.07	15.99
9	520.59	479.71	8.52	2.26	2.08	15.71
10	527.09	482.25	9.30	2.29	2.09	15.26
11	531.13	487.59	8.93	2.31	2.12	14.32
12	537.38	495.19	8.52	2.33	2.15	12.99
13	518.11	476.04	8.84	2.25	2.07	16.35
14	515.23	474.83	8.51	2.24	2.06	16.56
15	521.66	477.46	9.26	2.26	2.07	16.10
16	523.68	482.81	8.47	2.27	2.10	15.16
17	522.17	483.21	8.06	2.27	2.10	15.09
18	534.98	493.44	8.42	2.32	2.14	13.29
19	533.03	489.17	8.97	2.31	2.12	14.04
20	512.88	470.12	9.10	2.23	2.04	17.39
21	529.77	487.97	8.57	2.30	2.12	14.25
22	519.73	480.84	8.09	2.26	2.09	15.51
23	535.34	492.29	8.74	2.32	2.14	13.49
24	539.34	497.31	8.45	2.34	2.16	12.61
25	511.46	469.51	8.93	2.22	2.04	17.50
26	526.01	485.82	8.27	2.28	2.11	14.63
27	518.4	476.21	8.86	2.25	2.07	16.32
28	526.54	481.71	9.31	2.29	2.09	15.35
29	532.69	490.11	8.69	2.31	2.13	13.88
30	529.53	489.39	8.20	2.30	2.12	14.00
31	530.2	483.44	9.67	2.30	2.10	15.05
32	538.42	493.39	9.13	2.34	2.14	13.30

Table. 2 Physico-mechanical properties of Lower Fars fossiliferous limestone at different pH.

PH values	Sample No.	Volume of sample (cm <sup>3</sup> )	Dry weight of sample (g)	Saturated weight of sample (g)	Moisture Content (%)	Dry unit weight (g/cm <sup>3</sup> )	Saturated unit weight (g/cm <sup>3</sup> )	Uniaxial Compressive Strength (MPa)	Average of Uniaxial Compressive Strength (MPa)	Average of Modulus of Elasticity (MPa)
	1	230.4	480.95	0		2.09		20.23	20.60	693.02
	2	230.4	485.09	0		2.11		20.97		
1	3	230.4	465.04	516	10.96	2.02	2.24	4.02	4.06	350.23
	4	230.4	487.97	534	9.43	2.12	2.32	4.1		
3	5	230.4	478.59	522	9.07	2.08	2.27	5.24	5.10	518.96
	6	230.4	479.53	522	8.86	2.08	2.27	4.96		
5	7	230.4	478.28	518	8.30	2.08	2.25	7.07	6.44	638.83
	8	230.4	478.07	519	8.56	2.07	2.25	5.8		
7	9	230.4	479.71	519	8.19	2.08	2.25	8.15	8.43	748.93
	10	230.4	482.25	526	9.07	2.09	2.28	8.7		
9	11	230.4	487.59	530	8.70	2.12	2.30	7.9	7.75	778.74
	12	230.4	495.19	537	8.44	2.15	2.33	7.6		
11	13	230.4	476.04	519	9.02	2.07	2.25	6.98	6.81	665.33
	14	230.4	474.83	517	8.88	2.06	2.24	6.63		
13	15	230.4	477.46	522	9.33	2.07	2.27	5.36	5.62	480.87
	16	230.4	482.81	523	8.32	2.10	2.27	5.87		

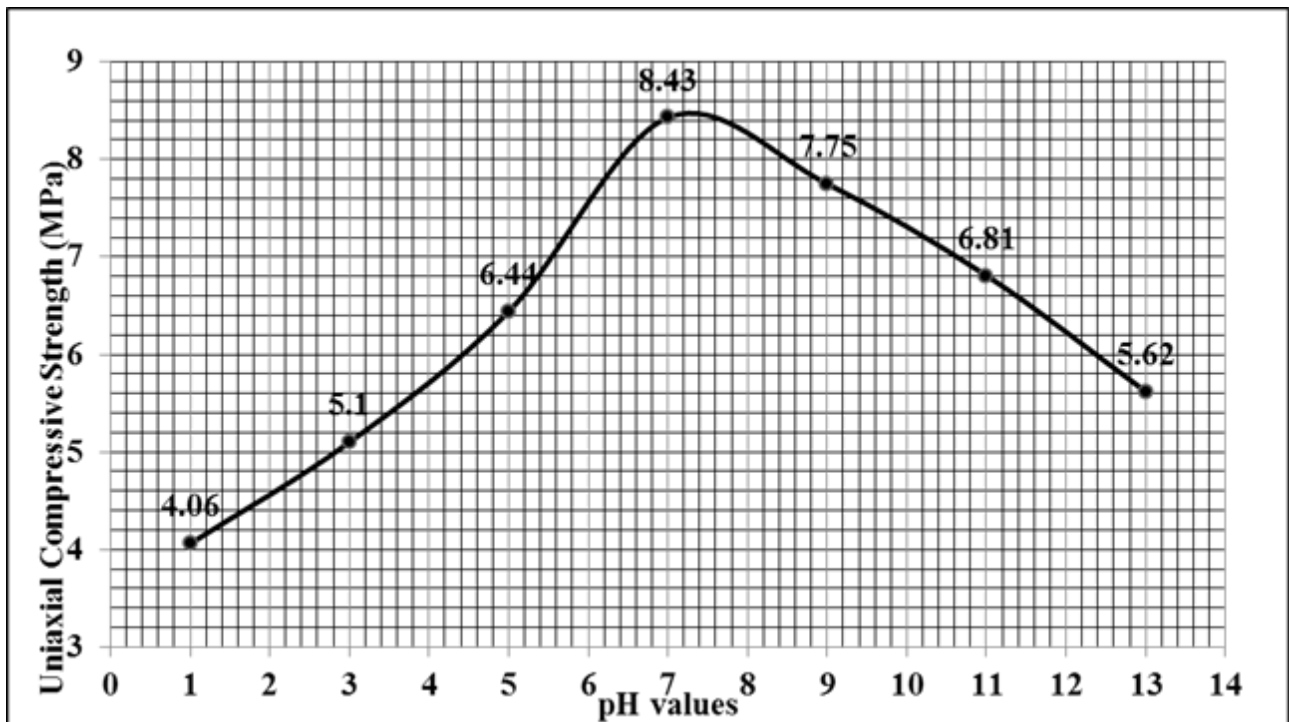


Fig. 5 Relation between uniaxial compressive strength and pH.

Reference [27] stated that the presence of liquids, substantially reduced the strength of the rocks. The reduction of the strength was attributed to the lowering of the surface free energy of the rock due to physical adsorption from the surrounding liquid.

The effect of moisture content on the uniaxial strength of chalk was investigated by [28]. According to their studies the compressive strength of chalk was less than 20 % of its dry strength in saturated condition. In the

present study the reduction of compressive strength is less than 80.29% of its dry strength at 1 pH and less than 72.72% of its dry strength at 13 pH.

Maximum Modulus of Elasticity was measured as 778.74 MPa at 9 pH. However, it shows a decreasing trend in acidic and alkaline condition, approximately similar to the other physico-mechanical properties (Table. 2 and Fig. 6). The Table. 3 and Fig. 7 show the results and relation between uniaxial compressive strengths and strains for specimens S3 to S16.

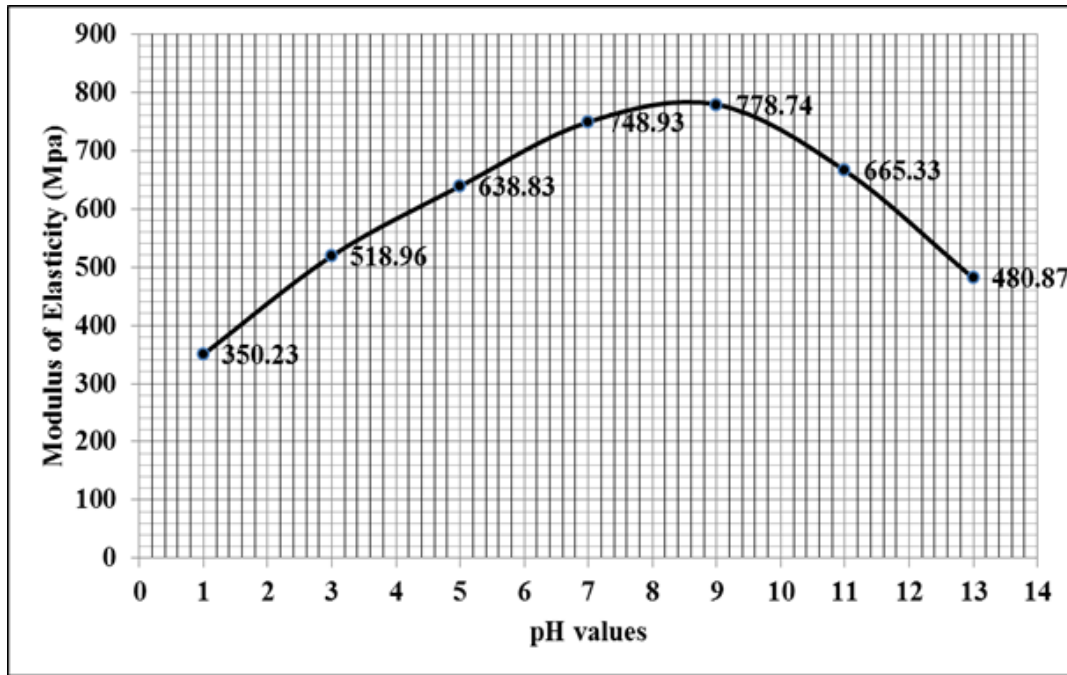


Fig. 6 Relation between Modulus of Elasticity and pH.

Table. 3 Axial stress versus strain at different pH solutions.

Strain %	Axial Stress (kN/m <sup>2</sup> )													
	pH 1		pH 3		pH 5		pH 7		pH 9		pH 11		pH 13	
	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-11	S-12	S-13	S-14	S-15	S-16
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0.1	52.0	47.7	234.1	104.1	108.4	130.1	290.5	86.7	177.8	216.8	130.1	65.0	65.0	173.4
0.2	142.9	117.0	506.8	173.3	268.6	433.2	736.4	173.3	606.4	649.7	303.2	194.9	129.9	454.8
0.3	354.8	246.7	900.1	346.2	571.2	908.7	1298.2	389.5	1254.9	1254.9	649.1	476.0	238.0	835.2
0.4	562.0	440.9	1383.3	562.0	968.3	1426.6	1902.1	734.9	1945.3	1945.3	1210.4	877.6	462.6	1296.9
0.5	798.9	708.2	1943.4	820.5	1524.5	1986.5	2599.8	1274.0	2677.5	2763.9	1813.8	1442.4	777.3	1774.9
0.6	1164.8	1022.5	2545.4	1108.8	2079.5	2545.4	3322.0	1945.7	3451.4	3580.8	2459.1	1984.5	1164.8	2286.5
0.7	1465.4	1374.9	3146.2	1456.7	2710.9	3146.2	4107.3	2637.7	4309.9	4357.3	3189.3	2663.5	1573.1	2801.4
0.8	1765.3	1748.1	3788.9	1829.9	3323.9	3831.9	4951.4	3358.3	5248.5	5295.8	4004.2	3276.5	2032.2	3358.3
0.9	2172.1	2137.7	4473.3	2279.6	4004.4	4430.3	5806.6	4172.2	6107.7	6026.0	4783.0	3987.2	2550.6	3957.1
1.0	2586.7	2578.1	5070.3	2711.3	4769.5	5027.3	6574.2	4984.4	6875.0	6960.9	5671.9	4597.7	3072.3	4511.7
1.1	3047.7	2979.0	5236.9	3176.5	5666.1	5537.4	7297.3	5794.9	7898.3	7615.0	6438.8	5236.9	3584.3	5073.8
1.2	3409.1	3387.7	5231.6	3645.0	6560.9	5703.3	8147.6	6689.6	7890.3	7607.3	6861.1	5831.9	4125.2	5450.3
1.3	3731.2	3769.8		4112.5	7068.4	5766.1	8139.3	7453.9			6982.7	6340.1	4626.6	5868.9
1.4	4022.7	4104.1		4557.7	7061.2	5760.2		8302.3			6975.6	6633.2	5049.8	5862.9
1.5	4018.7	4099.9		4925.0				8704.3				6626.5	5361.1	
1.6				4962.7				8695.4					5355.6	
1.7				4957.7										

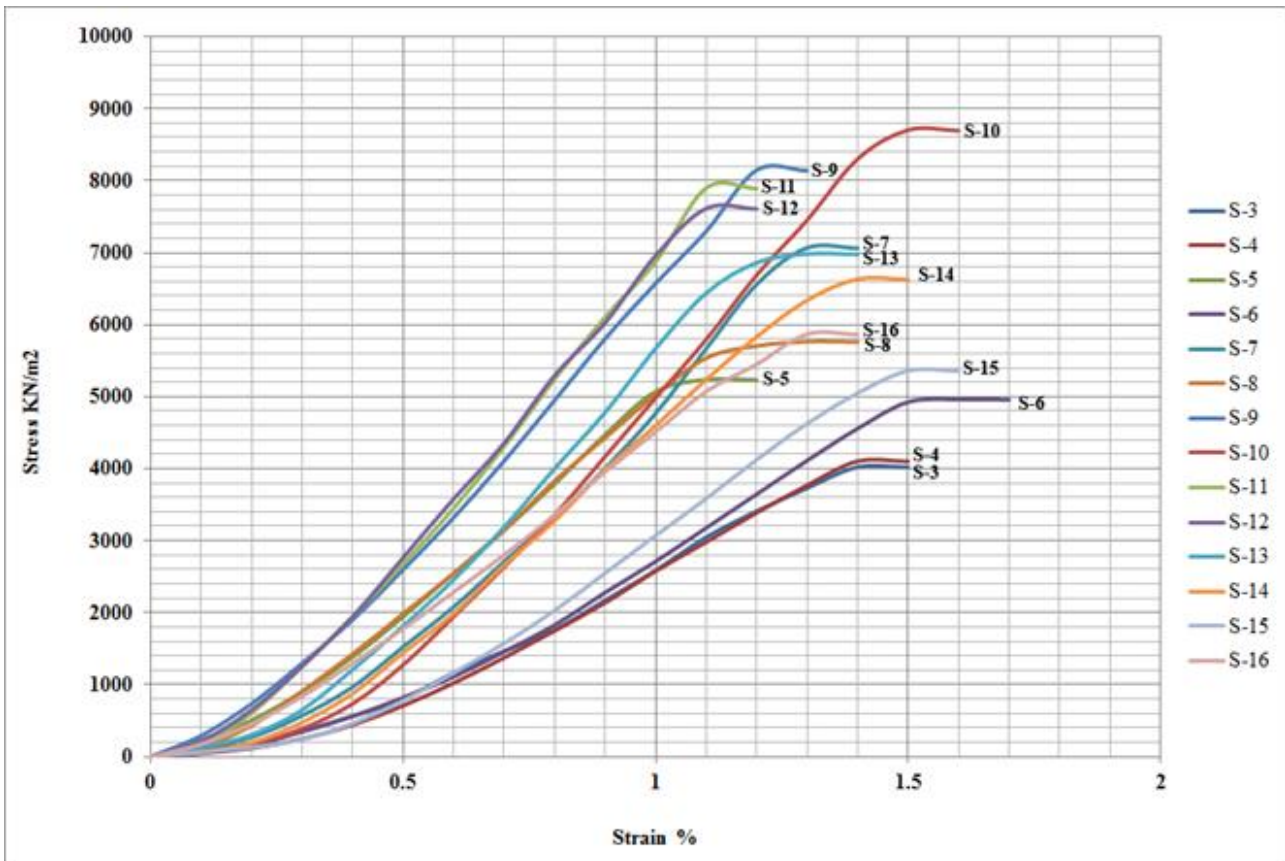


Fig. 7 Axial stress versus strain curves.

### 3- Point load strength test

The specimens were tested to determine the point load strength index by using a point load testing machine (Fig. 8). Point load strengths were calculated for both dry and saturated samples with pH values of (1, 3, 5, 7, 9, 11 and 13). Point load strength has a maximum value in dry condition which reaches the value 1.38 MPa. The maximum point load strength index in saturated condition was found at 7 pH with value of 0.45 MPa. However, it decreased under acidic and basic conditions with minimum value of 0.19 MPa at 1.0 pH and 0.21 MPa at 13 pH (Table. 4 and Fig. 9). The point load strength is less than 86.23% of its dry strength at 1 pH and less than 84.78% of its dry strength at 13 pH.



Fig. 8 Point load test

Table. 4 Physico-mechanical properties of Lower Fars fossiliferous limestone at different pH.

PH values	Sample No.	Volume of sample (cm <sup>3</sup> )	Dry weight of sample (g)	Saturated weight of sample (g)	Moisture Content (%)	Dry unit weight (g/cm <sup>3</sup> )	Saturated unit weight (g/cm <sup>3</sup> )	Point Load reading (KN)	Point Load strength Index I <sub>s</sub> (Mpa)	Point Load strength Index I <sub>s(50)</sub> (Mpa)	Average Point Load strength Index I <sub>s(50)</sub> (MPa)
0	17	230.40	483.21	0.00		2.10	0.00	4.98	1.70	1.75	1.38
	18	230.40	493.44	0.00		2.14	0.00	5.10	1.74	1.02	
1	19	230.40	489.17	533.00	8.96	2.12	1.92	0.92	0.31	0.18	0.19
	20	230.40	470.12	523.00	11.25	2.04	1.93	1.01	0.34	0.20	
3	21	230.40	487.97	529.00	8.41	2.12	1.96	1.26	0.43	0.25	0.22
	22	230.40	480.84	518.00	7.73	2.09	1.89	0.99	0.34	0.20	
5	23	230.40	492.29	533.00	8.27	2.14	1.96	1.19	0.41	0.24	0.26
	24	230.40	497.31	538.00	8.18	2.16	1.93	1.46	0.50	0.29	
7	25	230.40	469.51	511.00	8.84	2.04	1.96	2.49	0.85	0.50	0.45
	26	230.40	485.82	524.00	7.86	2.11	2.00	2.00	0.68	0.40	
9	27	230.40	476.21	516.00	8.36	2.07	1.88	1.31	0.45	0.26	0.24
	28	230.40	481.71	527.00	9.40	2.09	1.93	1.07	0.36	0.21	
11	29	230.40	490.11	531.00	8.34	2.13	1.91	1.06	0.36	0.21	0.23
	30	230.40	489.39	529.00	8.09	2.12	1.93	1.28	0.44	0.26	
13	31	230.40	483.44	523.00	8.18	2.10	1.97	1.02	0.35	0.20	0.21
	32	230.40	493.39	530.00	7.42	2.14	1.98	1.07	0.36	0.21	

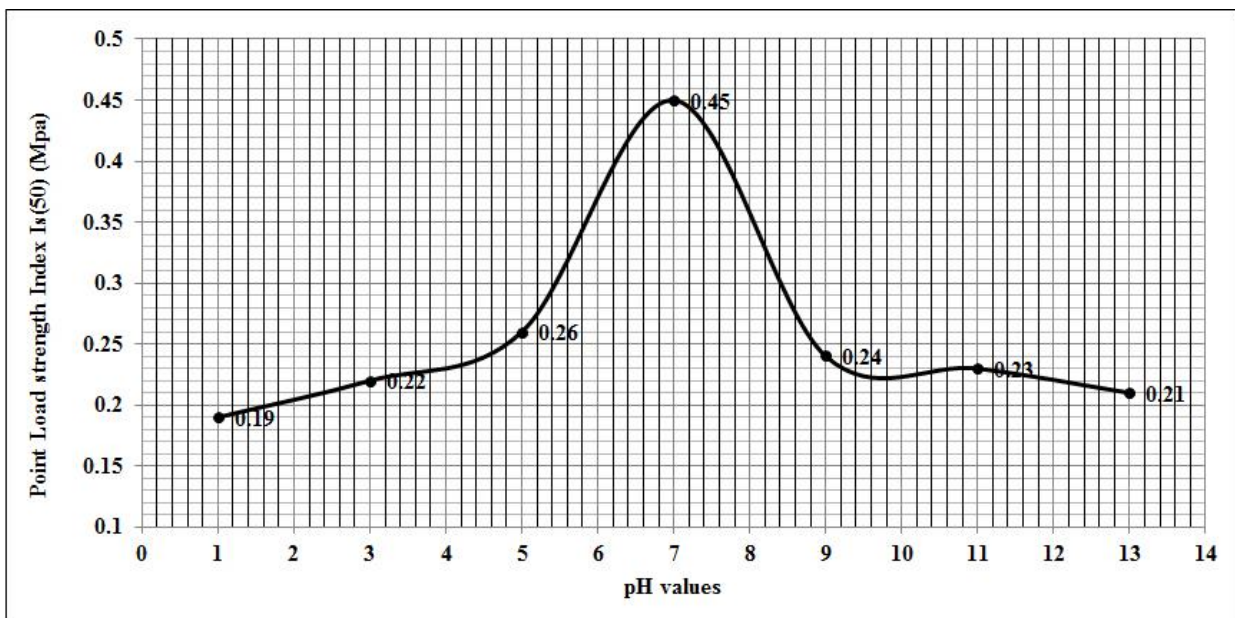


Fig. 9 Relation between pH and point load index.

## Conclusion

This study helps to estimate the behavior of limestone structures, which are exposed to different moisture conditions that are present in rainy atmosphere. Despite the laboratory tests carried out in this study are simplified compared with natural weathering processes in polluted atmospheric conditions, the results indicated that the limestone appears to be less resistant to the atmospheric factors. The following conclusions can be drawn:

1. The evaluation of the effective porosity has been identified into two ways: a. Immersion of samples in water till saturation point b. calculation of porosity based on thin sections under the microscope, using the point count method. In both methods the porosity ranges between 11% to 19%. This porosity makes the rock less resistance when exposed to weather conditions.
2. Estimation of Uniaxial compressive strength and point Load strength in dry and saturated conditions. The reduction of compressive strength is less than 80.29% of its dry strength at 1 pH and less than 72.72% of its dry strength at 13 pH. However, the reduction of point load strength is less than 86.23% of its dry strength at 1 pH and less than 84.78% of its dry strength at 13 pH.
3. The maximum compressive strength of 8.43MPa was found at pH value 7 in saturated conditions. This strength decreases in both acidic and alkaline conditions.
4. The maximum point load strength index in saturated condition was found to be at pH value 7 and it decreases under acidic and basic conditions.
5. The modulus of elasticity attained maximum value of 778.74 MPa at pH value 9. However, it shows a decreasing trend in acidic and basic conditions.

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