CORRELATIONS FOR LIMESTONE PROPERTIES USED IN CULTURAL HERITAGE MONUMENTS IN MALTA

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Abstract. The aim of this paper is to identify mathematical correlations between the physical and geochemical properties of first quality limestone extracted from the Lower Globigerina Member. Based on published results, it can be confirmed that very strong correlations exist for (i) apparent density and uniaxial compressive strength when limestone is in a saturated condition, and (ii) ultrasonic pulse velocity – both perpendicular and parallel to the bedding plane – and uniaxial compressive strength when limestone is in either oven-dried or saturated conditions. Stronger correlation is present with respect to apparent density when limestone is oven-dried and color. The correlations for color and (a) ferric oxide, and (b) loss-on-ignition are also strong.

Key words: limestone, Lower Globigerina Limestone, franka, Neolithic architecture, Malta.

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1. INTRODUCTION

Malta has one of the highest densities of buildings of cultural significance in the world. A leading contemporary archeologist labelled it an archeological paradise [1]. In terms of geological timescale, the Lower Globigerina member – the oldest of the three-tier Globigerina Limestone Formation – is Aquitanian [2–5]. The limestone from this member is known as Lower Globigerina Limestone (LGL), referred to in the local building trade as ‘franka’. LGL outcrops over approximately two-thirds of the island and, as such, is the main building material used for construction on the island [6]. The built heritage of the island – from Neolithic temples (some included in the UNESCO World Heritage List [7]) to Mdina (the capital from antiquity until the late sixteenth century) and Valletta (the capital and a UNESCO World Heritage Site [8]) – is erected in LGL, the largest number of sites dating to the period of the Hospitaller Order of St John of Jerusalem [9]. In 2017, a proposal was filed to nominate LGL as a Global Heritage Stone Resource [10].

The first comprehensive study of the physical, textural, mineralogical and geochemical properties of LGL, completed in 1993, was undertaken at the University of Leicester through funding provided by the Oil Exploration Directorate, Office of the Prime Minister, Malta [11]. The main findings of this study were later published [12, 13]. Prior analytical studies on particular characteristics of LGL were undertaken by the Building Research Station (BRS), Watford, UK, and the University of Malta.

At the BRS, a study was carried out on the relation between the durability of the Malta's limestone and the laboratory-measured properties and efficacy of silicone treatments [14]. Another study was conducted on the properties and behavior of local limestone [15]. All studies undertaken at the University of Malta were undergraduate dissertations. They addressed the engineering properties of the rocks and soils of Malta [16] and the mechanical and physical properties [17] – notably shear strength [18] and elastic constants [19] – of Globigerina Limestone.

Two main lithotypes of LGL are utilized in the building industry; the first is more durable than the second. The former is characterized by an acid-insoluble residue of < 5%; the latter – locally known as 'soll' – includes higher percentage of quartz fragments, K-feldspar and clays [11, 13]. Addressing the first quality lithotype, the aim of this article is to study correlations between the physical and geochemical parameters.

2. MATERIALS AND METHODS

Based on published experimental data [12, 13, 20], the physical and geochemical properties of eight 100 × 100 × 100 mm samples (S1 to S8) are given in Table 1 and Table 2, respectively. To assess the physical properties, the following testing methods were used: (i) apparent density (ρ), (ii) uniaxial compressive strength (UCS) using the Avery-Denison model; (iii) ultrasonic pulse velocity (UPV) using the PUNDIT model; and (iv) color (Eab) using (EEL) Abridged Reflectance Spectrometer model. For methods (i) and (ii), tests were undertaken on samples that had been oven-dried (temperature 105 +/– 5°C) and saturated (fully submerged for 24 hours). With respect to (ii), a constant loading rate of 0.15 N/mm^2 was applied perpendicular to the bedding plan. For (iii), samples were oven-dried only and readings were recorded perpendicular and parallel to the bedding plane. For the bulk chemistry, an ARL 8420+ X-ray fluorescence (XRF) spectrometer was used on pressed powder pellets [21]. The loss on ignition (LOI) was used to calculate the organic and carbonate content [22]; all values were < 44%, the theoretical value for pure $CaCO_3$.

Table 1

Physical properties

Sample Ref. no.	ρ (kg/m³)		UCS (N/mm²)		UPV (km/s)		Eab
	(oven-dried)	(saturated)	(oven-dried)	(saturated)	perpendicular	parallel	
S01[1, 2]	1778	2016	30.18	15.42	03.06	02.99	03.36
S02[1]	1717	1949	15.58	08.27	02.69	02.71	03.14
S03[1]	1784	1975	27.88	17.43	03.22	03.11	03.44
S04[1]	1787	1999	22.54	16.95	03.07	02.94	03.47
S05[3]	1718	1939	17.12	10.78	02.62	02.68	03.45
S06[3]	1725	1992	29.52	18.59	03.09	03.00	03.16
S07[3]	1693	1955	24.84	15.22	03.01	02.88	05.51
S08[3]	1799	2021	25.04	14.72	03.00	02.96	03.84

[1] Reproduced in [13], [2] in [12], and [3] in [20].

Table 2

Geochemical analysis (%)

Sample Ref. no.	XRF Analysis										LOI
	CaO	SiO_2	Al_2O_3	MgO	Fe_2O_3	K_2O	P_2O_5	TiO_2	Na_2O	MnO	
S01[1, 2]	49.67	06.77	01.06	01.14	00.60	00.36	00.21	00.15	00.04	00.03	41.72
S02[1]	50.22	04.27	00.43	00.72	00.32	00.19	00.19	00.07	00.07	00.04	42.81
S03[1]	49.77	04.45	00.43	01.12	00.37	00.19	00.58	00.08	00.07	00.03	42.57
S04[1]	48.79	05.09	00.51	01.17	00.63	00.25	01.08	00.08	00.14	00.04	42.01
S05[3]	50.08	04.95	00.68	01.15	00.34	00.25	00.24	00.09	00.06	00.03	42.52
S06[3]	52.86	03.28	00.97	00.84	00.36	00.29	00.12	00.09	00.00	00.00	42.47
S07[3]	52.49	03.06	00.91	00.85	00.31	00.26	00.15	00.08	00.05	00.00	42.64
S08[3]	50.14	06.38	01.57	00.91	00.91	00.53	00.29	0.18	00.01	00.00	40.40

[1] Reproduced in [13], [2] in [12], and [3] in [20].

With the exception of color analysis, all other physical experiments were carried out at the Department of Civil Engineering of the Faculty of Architecture and Civil Engineering, the forerunner of the Faculty for the Built Environment, University of Malta. Color analysis and XRF were undertaken at the Department of Geology, now integrated in the School of Geography, Geology and the Environment, University of Leicester.

3. RESULTS AND DISCUSSION

The various combinations possible from the data for ρ, UCS and UPV were plotted and the best fit curve – all second-order polynomials – was derived for each using Excel (Microsoft®, Excel® 2019). The respective coefficient of determination, R-squared (R^2), where R is the coefficient of correlation, was computed. Whilst R measures the strength of the relationship between two variables, R^2 measures the amount of variation in the dataset. The R^2 and R between physical parameters and color are given in Table 3 and Table 4, respectively. The resulting coefficients between geochemical parameters and color are given in Table 5.

R^2 is a measure of how well a linear regression model fits the dataset; it is a measure of goodness of fit. The closer the value is to 1, the better the fit. Given that an R^2 is 0.64 (*i.e.*, the regression model explains 64% of the variability in the data), the R value of 0.8 or 80% is a strong coefficient of correlation; the model for a set of given parameters is a moderately accurate fit. Thus, the range for the model considered a moderate fit to be $R^2 \geq 0.64$. A more accurate fit is $R^2 \geq 0.75$. Thus, only correlations between the relevant parameters at such values are plotted in Fig. 1 and Fig. 2. The equation for the curve that best fits the dataset is included. Although given in quadratic functions, the approximations for Fig. 1a and Fig. 2c are almost linear since the x^2 coefficient is nearly 0.

Table 3

Coefficient of determination (R^2) and coefficient of correlation (R) between physical parameters

	R^2	R
ρ (oven-dried) and UCS (oven-dried)	0.16	0.40
ρ (saturated) and UCS (saturated)	0.75	0.87
UPV (perpendicular) and ρ (oven-dried)	0.29	0.54
UPV (perpendicular) and ρ (saturated)	0.58	0.76
UPV (parallel) and ρ (oven-dried)	0.38	0.62
UPV (parallel) and ρ (saturated)	0.63	0.79
UPV (perpendicular) and UCS (oven-dried)	0.78	0.88
UPV (perpendicular) and UCS (saturated)	0.85	0.92
UPV (parallel) and UCS (oven-dried)	0.84	0.92
UPV (parallel) and UCS (saturated)	0.83	0.91

Table 4

Coefficient of determination (R^2) and coefficient of correlation (R) between physical parameters and color

	ρ (kg/m^3)		UCS (N/mm^2)		UPV (km/s)	
	(oven-dried)	(saturated)	(oven-dried)	(saturated)	perpendicular	parallel
R^2	0.89	0.04	0.37	0.32	0.18	0.21
R	0.94	0.20	0.61	0.57	0.42	0.46

Table 5

Coefficient of determination (R^2) and coefficient of correlation (R) between geochemical parameters and color

	XRF Analysis										LOI
	CaO	SiO_2	Al_2O_3	MgO	Fe_2O_3	K_2O	P_2O_5	TiO_2	Na_2O	MnO	
R^2	0.51	0.60	0.27	0.32	0.64	0.42	0.18	0.43	0.01	0.29	0.64
R	0.71	0.77	0.52	0.57	0.80	0.65	0.42	0.66	0.10	0.54	0.80

The effect of water on ρ and UCS is significant (Table 1). The difference in ρ is indicative of the porosity of LGL; water uptake ranges from 11 to 15%. The presence of water in pores leads to a reduction of UCS in the range of 25 to 50%. Given that UPV values were obtained for oven-dried samples, they cannot be compared and contrasted with saturated ones; UPV values in saturated conditions are higher than oven-dried ones [23]. Also, as noted by Vasanelli *et al.* [23], an increase in clay content leads to a decrease in UPV [24–27].

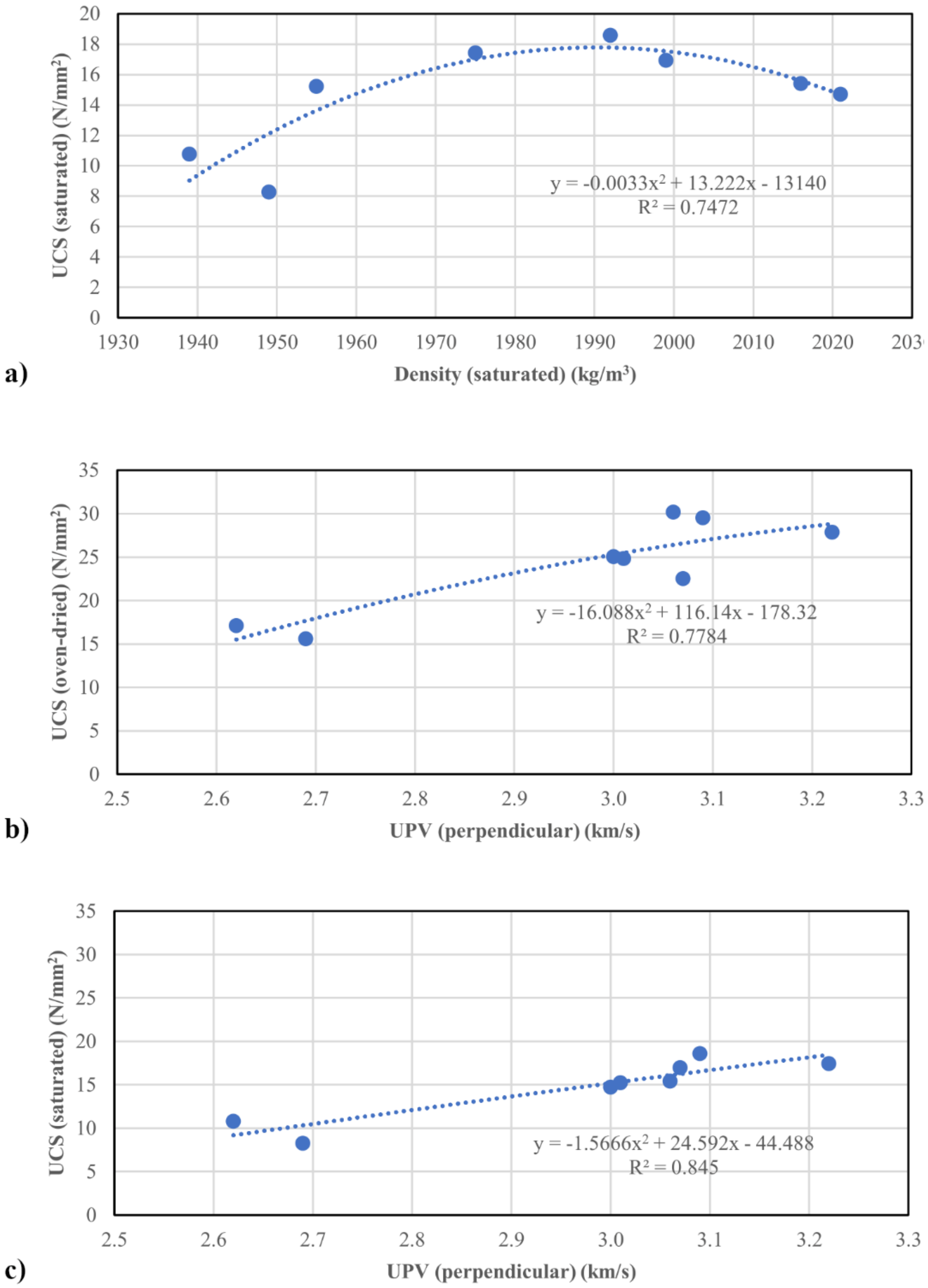
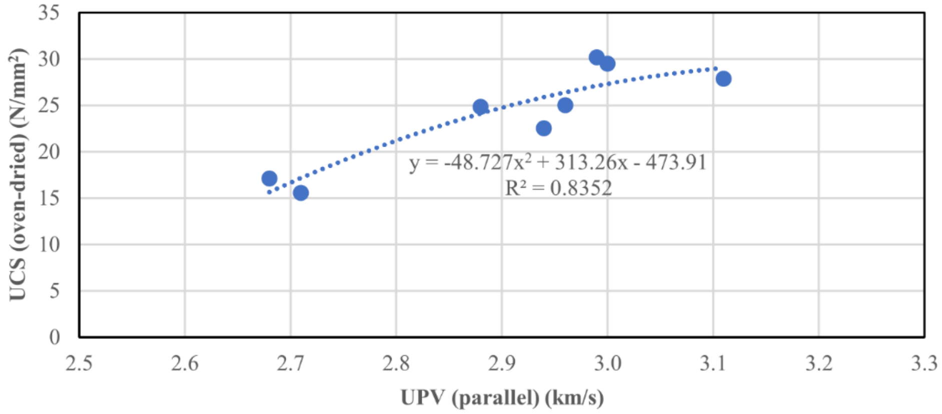
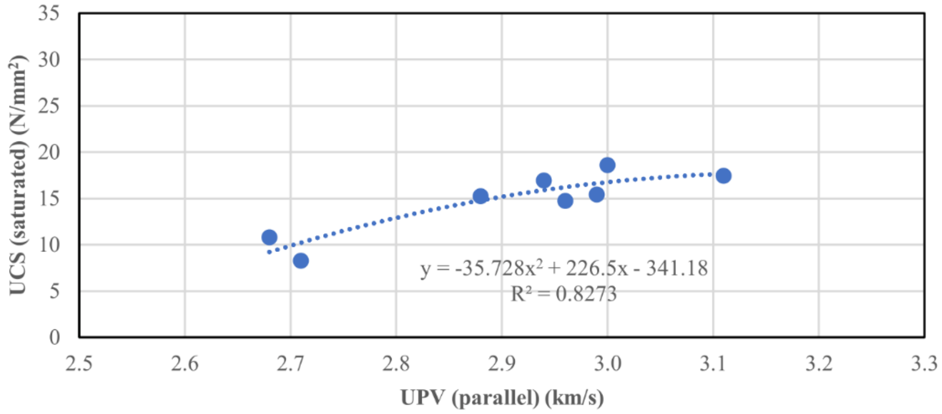


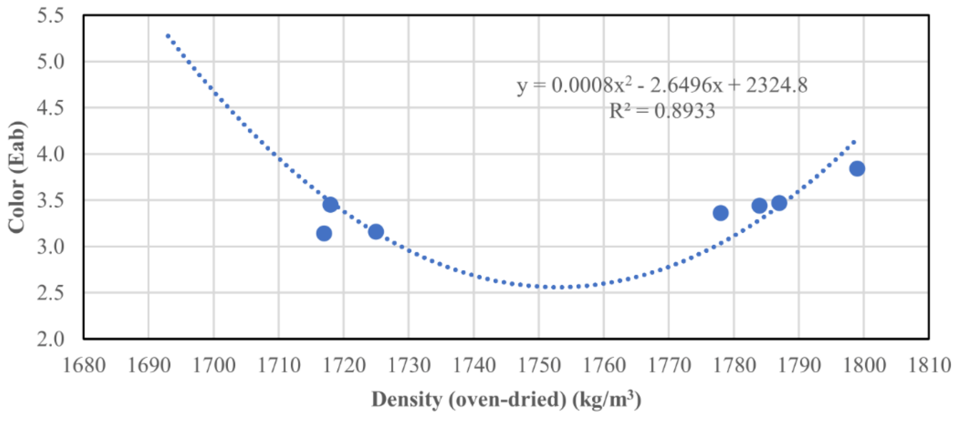
Fig. 1 – Correlation between: a) ρ (saturated) and UCS (saturated), b) UPV (perpendicular) and UCS (oven-dried) and c) UPV (perpendicular), and UCS (saturated).



a)



b)



c)

Fig. 2 – Correlation between: a) UPV (parallel) and UCS (oven-dried), b) UPV (parallel) and UCS (saturated), and c) ρ (oven-dried) and color.

With respect to the geochemical parameters, at $R^2 = 0.64$, strong correlations exist between Eab and (i) Fe_2O_3 ($R = 0.80$), and (ii) LOI ($R = 0.80$). The strongest correlation ($R = 0.94$) was found, and a more reliable regression model (with higher accuracy) was determined, between Eab and ρ (oven-dried) ($R^2 = 0.89$; Fig. 2c). The correlation between UCS and ρ is present on the samples in a saturated condition ($R^2 = 0.75$; Fig. 1a) is stronger ($R = 0.87$) than that for the above stated geochemical parameters. The overall, strong and reliable correlation between UCS and UPV supports the findings on Lecce stone [23]; in all scenarios, R is approximately 90% and thus the model fits the dataset well. For UPV (perpendicular) and UCS (oven-dried), the model explains 78% of the variability in the data ($R^2 = 0.78$) (Fig. 1b). The correlation of UPV (perpendicular) and UCS (saturated) is even stronger ($R = 0.92$; Fig. 1c). For data recorded parallel to the bedding plan, there is a very strong correlation between the UCS of limestone at oven-dried and saturated conditions; at $R = 0.92$ (Fig. 2a), the former was found to be marginally better than the latter ($R = 0.83$; Fig. 2b).

4. CONCLUSIONS AND FINAL COMMENTS

At $R \geq 0.80$, a strong correlation exists between any two variables. Thus,

1. with respect to the geochemical parameters, strong correlations exist between Eab and (i) Fe_2O_3, and (ii) LOI;
2. with respect to the physical parameters, stronger correlations exist between (i) ρ and UCS when the limestone was in saturated state, (ii) UPV and UCS for the possible combinations: perpendicular and parallel for oven-dried and saturated conditions; and
3. the strongest correlation exists between Eab and ρ (oven-dried).

Due to the small number of samples, the correlation coefficients are only indicative; the larger the number of samples analyzed, the higher the accuracy of the indication of degree of fit of the regression model.

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REFERENCES

1. A. Bonanno, J. Bartolo and M. Mintoff, *Malta: an archaeological paradise*, M. J. Publications, Valletta, 1991.
2. R. Felix, *Oligo-Miocene Stratigraphy of Malta and Gozo*, H. Veenman and B.V. Zonen, Wageningen, The Netherlands, 1973.
3. M.R. House, K.C. Dunham and J.C. Wigglesworth, *Geology of the Maltese Islands*, in: H. Bowen-Jones, J.C. Dewdney and W.B. Fisher (Eds.), Malta: Background for development, University of Durham: Newcastle, UK, 1961, pp. 24–33.
4. H.M. Pedley, M.R. House, and B. Waugh, *The geology of the Pelagian Block: The Maltese Islands*, in: A.E.M. Nairn, W.H. Kanes, F.G. Stehli (Eds.), *The Ocean Basins and Margins*, Springer, Boston, MA, USA, 1978; Volume 4B, pp. 417–433.

5. H.M. Pedley, M.R. House, and B. Waugh, *The geology of Malta and Gozo*, Proceedings of the Geologists' Association, **87**, 325–341 (1976).
6. Office of the Prime Minister, *Geological map of the Maltese Islands: Sheet 1 – Malta*, Oil Exploration Directorate, Malta, 1993.
7. UNESCO, *World Heritage List: Megalithic Temples of Malta*. http://whc.unesco.org/en/list/132. Accessed on 30.07.2023.
8. UNESCO, *World Heritage List: City of Valletta*. http://whc.unesco.org/en/list/131. Accessed on 30.07.2023.
9. J. Quentin Hughes, *The building of Malta during the period of the Knights of St John of Jerusalem*, Alec Tiranti, London, 1967.
10. J. Cassar, A. Torpiano, T. Zammit and A. Micallef, *Proposal for the nomination of Lower Globigerina Limestone of the Maltese Islands as a "Global Heritage Stone Resource"*, Journal of International Geoscience, **40**, *3*, 221–231 (2017).
11. L. Bianco, *Some factors controlling the quality of Lower Globigerina building stone of Malta*, Unpublished M.Sc. dissertation, University of Leicester, Leicester, 1993.
12. L. Bianco, *Techniques to determine the provenance of limestone used in Neolithic architecture of Malta*, Romanian Journal of Physics, **62**, *1–2*, 901 (2017).
13. L. Bianco, *Geochemistry, mineralogy and textural properties of the Lower Globigerina Limestone used in the built heritage*, Minerals, **11**, 740 (2021).
14. Building Research Station, *Maltese Limestones: Relation of durability to laboratory-measured properties and efficacy of silicone treatments*, Unpublished note no. C965, Department of Scientific and Industrial Research, BRS, Watford, UK, 1963.
15. Building Research Station, *The Maltese Islands: Properties and behavior of local limestone*, Unpublished internal note 6, Department of Scientific and Industrial Research, BRS, Watford, UK, 1964.
16. S. Bonello, *Engineering properties of rocks and soils of the Maltese Island*, Unpublished B.E.&A. (Hons) dissertation, University of Malta, Msida, 1988.
17. J. Cachia, *The mechanical and physical properties of the Globigerina Limestone as used in local masonry construction*, Unpublished B.E.&A. (Hons) dissertation, University of Malta, Msida, 1985.
18. J. Saliba, *The shear strength of the Globigerina Limestone*, Unpublished B.E.&A. (Hons) dissertation, University of Malta, Msida, 1990.
19. D. Xuereb, *Elastic constants of Globigerina Limestone*, Unpublished B.E.&A. (Hons) dissertation, University of Malta, Msida, 1991.
20. L. Bianco, *Limestone replacement in restoration: the case of the church of Santa Maria (Birkirkara, Malta)*, International Journal of Conservation Science, **8**, *2*, 167–176 (2017).
21. G. Fitton, *X-ray fluorescence spectrometry*, in: R. Gill (Ed.), Modern analytical geochemistry: an introduction to quantitative chemical analysis for earth, environmental and material scientists, Longman: Harlow, UK, 1997, pp. 97–115.
22. W.E. Dean, *Determination of carbonate and organic matter in calcareous sediments and sedimentary rocks by loss on ignition: Comparison with other methods*, Journal of Sedimentary Petrology, **44**, *1*, 242–248 (1974).
23. E. Vasanelli, D. Colangiuli, A. Calia, M. Sileo and M.A. Aiello, *Ultrasonic pulse velocity for the evaluation of physical and mechanical properties of a highly porous building limestone*, Ultrasonics, **60**, 33–40 (2015).
24. D. Han, A. Nur and D. Morgan, *Effects of porosity and clay content on wave velocities in sandstones*, Geophysics, **51**, 2093–2107 (1986).
25. T. Klimentos, *The effects of porosity-permeability-clay content on the velocity of compressional waves*, Geophysics, **56**, 1930–1939 (1991).
26. H. Karakul, R. Ulusay, *Empirical correlations for predicting strength properties of rocks from P-wave velocity under different degrees of saturation*, Rock Mechanics and Rock Engineering, **46**, 981–999 (2013).
27. M.S. King, *Wave velocities in rocks as a function changes in overburden pressure and pore fluid saturants*, Geophysics, **31**, 50–73 (1966).