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MINERALOGY AND GEOCHEMISTRY OF BLUE PATCHES OCCURRING IN THE *GLOBIGERINA* LIMESTONE FORMATION USED IN THE ARCHITECTURE OF THE MALTESE ISLANDS

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Abstract

The stratigraphy of the Maltese Archipelago is composed of Oligo-Miocene industrial minerals of shallow marine origin. The Lower *Globigerina* Limestone Member, the earliest of the three members forming the *Globigerina* Limestone Formation, had been used as the main building material of the islands since the Neolithic Period. A bluish coloured lithostratigraphic bed, known in the quarry industry as *sol ikħal*, occurs within this unit. Furthermore, occasionally, large blue patches occur in this member. They were first studied by John Murray whose research was published towards the end of the nineteenth century. This article analyses the mineralogy and geochemistry of *sol ikħal* and these patches applying XRF and XRD. Whilst the results for the blue patches corroborate and refine the findings of Murray, they differ from *sol ikħal.* Their SiO_2 content is $> 10\%$; for *sol ikħal* it is $< 10\%$.

Key words: *Globigerina* Limestone, sol ikħal, John Murray, Maltese Archipelago, Malta

Introduction. The Maltese Archipelago is situated nearly at the centre of the Mediterranean Sea. Malta, the largest southerly island, is 27 km in length compared to the next larger northerly island of Gozo which is 14.5 km. In between there is the third island of Comino measuring 2.5 km. There are a number of much smaller, uninhabitable, islands. The archipelago, having a superficial area of circa 316 km^2, is located 93 km south of Ragusa Peninsula in Sicily and 288 km north of Libya.

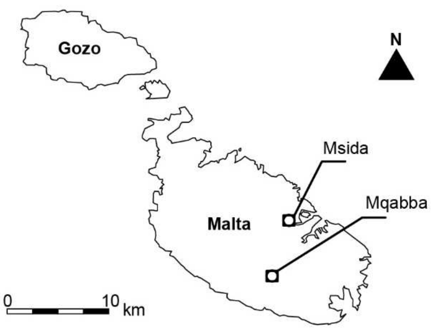
The archipelago is situated on the eastern edge of the North African Pelagian Shelf which runs from the coast of Tunisia to the west of the Ionian Sea and from the coast of Libya to the south of Sicily. It is composed of mid-Tertiary carbonate rocks of shallow marine origin laid in a simple succession of the five Oligo-Miocene frequently fissured formations.

The Lower Globigerina Limestone Member has been extensively quarried for use in the building tradition since the Neolithic Period. It provides the islands with the characteristic honey-coloured dimension stone, the medium in which rich architectural legacy is realized. This member has a distinctively blue coloured bed of *sol ikħal* (*ikħal* being the Maltese word for 'blue'). Occasionally, similar coloured, large patches also occur in this member. The scope of this paper is to investigate whether the mineralogy and geochemistry of *sol ikħal* differ from the blue patches (Fig. 1(a), (b)). These patches were first studied by MURRAY [1], an authority on oceanic sedimentation and the father of modern oceanography.

Geological setting. Stratigraphically, the islands are, in geological timescale starting from the earliest, composed of the following formations: Lower Coralline Limestone, Globigerina Limestone, Blue Clay, Greensand and the Upper Coralline Limestone. The succession is capped by Quaternary deposits [2]. Malta and Gozo are the only islands which display the whole stratigraphic sequence. A historical overview of the industrial minerals on the island is given by BIANCO [3]. The general literature on the lithostratigraphy of the islands dates back to the nineteenth century [1, 4–6] and remained essentially reiterated in basic geological texts [7–9]. It was refined in the later part of the twentieth century [10–13]. A new lithostratigraphical and palaeoenvironmental interpretation of the Coralline Limestone formations is given by PEDLEY [14].

The Lower Coralline Limestone Formation is composed of massive bedded, pale grey to white, algal limestone, commonly detrital and rich in fauna. The Globigerina Limestone Formation is the one which outcrops most in Malta and Gozo. It consists of yellow to off-white, fine grained, massive bedded limestone with abundant Globigerinidae and planktonic foraminifera [1, 13, 15, 16]. Lithologically the formation is subdivided into three well discernible subdivisions: Lower Globigerina Limestone (cream coloured, dominantly planktonic foraminiferal), Middle Globigerina Limestone (whitish in colour, dominantly coccolithic), and Upper Globigerina Limestone (cream coloured, dominantly planktonic foraminiferal) [17]. The transition is marked by phosphorite horizons at the top of the lower and middle subdivisions, the lower main conglomerate bed C1 and the upper main conglomerate bed C2, respectively. These horizons have a mean thickness of about 0.5 m and mark an interruption in sedimentation [17]. The calcareous plankton bio-chronostratigarphy of the Lower Globigerina Limestone Member was the subject of recent research [18].

The blue patches contain pyrite, a mineral otherwise absent in the Lower Globigerina Limestone Member, and "often filling the foraminifera and forming

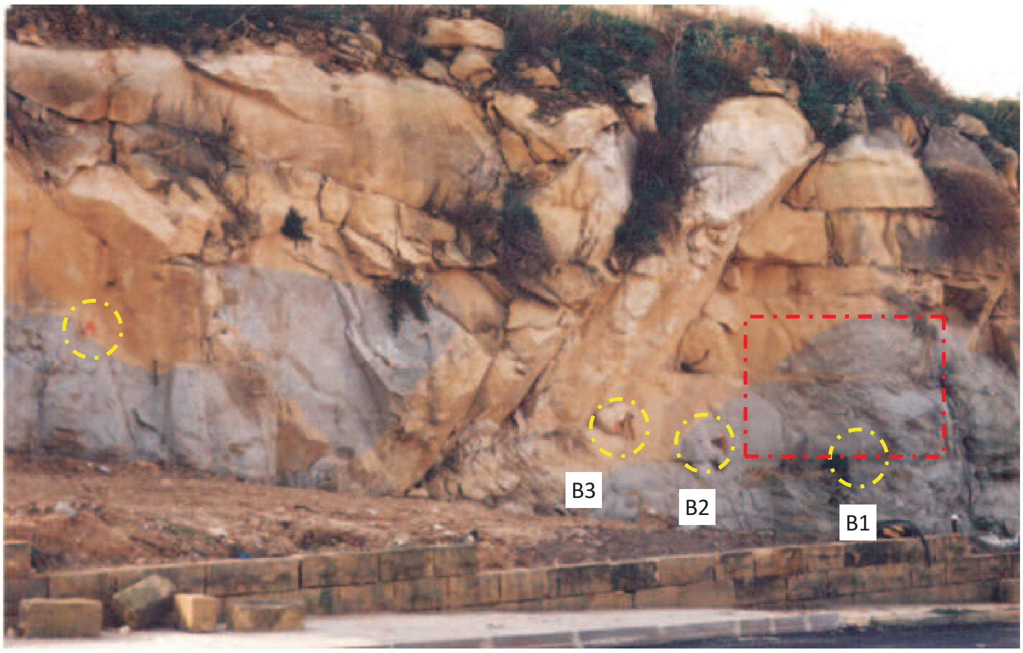


(a)

(b)

(c)

Fig. 1. (a) The habitable islands of the Maltese Archipelago; (b) orthophoto (© Planning Authority, Malta) showing the location (circled in red) of the blue patches in the Lower Globigerina Limestone Member at Msida; (c) the site (area outlined in red) from where the respective samples B1 to B3 were extracted



(a)

(b)

Fig. 2. (a) The location of samples (circled) is noted; (b) position of sample B1 is visible (see arrow); the scale purposes, the head of geological hammer is 160 mm long

Table 1

Chemical analysis of dried samples [1]

	Host rock	*Blue patch*
Carbonate of lime ($CaCO_3$, Fe_2O_3 and Al_2O_3)	81.37	78.39
Phosphate of lime (Ca_32PO_4)	03.57	02.70
Magnesium carbonate ($MgCO_3$)	01.63	00.44
Calcium sulphate ($CaSO_4$)	00.06	00.33
Insoluble residue in dilute HCl (1 in 10)	12.88	17.87
Total	*99.51*	*99.73*

casts of the shells" [1]. The results documented by Murray are summarized in Table 1. Two samples were collected from the same horizon, one from the blue patch and the other from the host rock. The insoluble residue in the latter consists of ferric oxide, alumina, silica, and a small quantity of lime, but no phosphoric acid or sulphur compound. The silica seems to occur in combination with the alumina. In the former, the insoluble residue consists of iron (mainly ferrous), abundant alumina, small amount of lime, and traces of sulphur but no phosphoric acid. The iron seems to be combined as silicate due to difficulty in decomposing the compound, as the colour remained unchanged even after repeated application of HCl, and subsequent evaporation. "The sulphur is present as sulphide which on oxidation in the presence of bases resulted in Fe_2O_3, while calcium sulphate would probably be formed" [1].

The Blue Clay Formation consists of alternating pale grey and dark marls with lighter bands containing higher carbonate content [13]. This content is never over 30% [1]. It is Serravallian in age [19] except for the upper levels which are Tortonian. Being an impervious layer, this formation forms the base of the perched aquifer [20]. The Greensand Formation, Messinian in age, is composed of poorly cemented bioclastic, glauconitic limestone [13, 14]. Each of the Coralline Limestone Formations is subdivided into four members [2]. The Greensand Formation is included in the Għajn Melel Member, the earliest member of the Upper Coralline Limestone Formation [2].

Materials and methods. The dominant mineralogy of the Lower Globigerina Limestone Member is calcite with minor inclusions of quartz, K-feldspar, muscovite, kaolinite, illite, smectite and glauconite. Four samples, all analysed in [21] but never published, are as follows: B1 to B3 obtained from a blue patch along the road to Msida cut through the Lower Globigerina Limestone Member (UTM ED50 coordinates: 453212E, 3972483N) (Fig. 1, 2) and B4, a sample of *sol ikħal* from the *Piccolo Fewda* quarry at Mqabba (UTM ED50 coordinates: 452215E, 3967496N) (Fig. 1(a)) which sample was identified by late Salvatore Bondin, the owner and operator of the quarry. The B4 sample is from an inferior quality of Lower Globigerina Limestone Member which is suitable only for use in foundations and as fill (Salvatore Bondin, personal communication). At the site in Msida, the sheared plane within the blue patch is marked with yellow bands on either side. Sample '4' shown in Fig. 2 (a) was not included because a fault is present between the location of this sample and B1 to B3. With respect to sample B4, no detail on the horizon from where it was extracted is available.

Petrographical microscopy was used for thin-section analysis to investigate the texture, porosity and permeability. Chemical analysis was determined through X-ray fluorescence analysis (XRF). The pressed powder pellets were analysed by ARL 8420+ X-ray fluorescence spectrometer. The insoluble residue (IR) determined the non-carbonate fraction present. Its mineralogical composition was analyzed through X-ray diffraction (XRD) using Philips PW1729 X-ray generator. XRD was also used to determine the mineralogy of the whole rock and the clay fractions. To enhance the d001 peaks, an oriented mount technique was used to prepare the clay minerals [22].

Results and discussion. The XRF analysis of the samples is given in Table 2 whilst the summary of the mineral phases identified through XRD are listed in Table 3.

The fabric of samples B1 to B3 is mud-supported, biomicritic wackestone. Fine crystalline sparry calcite weakly cements the allochems and partially fills the inter-particle space within the unfilled, undamaged chambers. Porosity is of intra-particle type and areas of high permeability are well-defined. Burrows cut through the fabric. Their infill is wackestone of low porosity.

The main components are unbroken pelagic foraminifera (mostly Globigerina), quartz, glauconite, and iron oxide. The minerals and their corresponding chemistry are well identified. Glauconite was not detected through XRD. Quartz grains, rounded or elongated (mean size 25 µm; maximum 70 µm) account for 10% of the rock matrix. Burrow infill has more quartz but less glauconite than the host rock. They occur in a less random distribution than glauconite (maximum 100 µm), a mineral exhibiting breakdown. Iron oxide mineral (maximum grain size 30 µm) occurs in clusters.

Sample B1 has two defined bands: yellow and blue. The unbroken Globigerina content in the yellow and blue sections is 15% and 5%, respectively. In the former, quartz content is $< 5\%$ (maximum size 40 µm) whilst in the latter is $> 5\%$ (maximum size 65 µm) and less randomly distributed. The yellow coloured band has a 500 µm diameter compact, low porosity, burrow which is infilled with wackestone. In sample B4, a packstone with 25% of the host rock comprises undamaged, unfilled Globigerina. Quartz grains (30 µm maximum size) are not well-sorted and glauconite grains (maximum 25 µm) are scarce. Weathering iron oxide grains (15 µm in diameter) are locally visible. A burrow (1 mm in diameter) cuts across the fabric B4. Burrow infill is wackestone; maximum 120 µm diameter inter-particle pores are present. The unbroken allochems in the burrow are partly

T a b l e 2

XRF analysis of limestone samples

Oxides	B1	B2	B3	B4
SiO_2	17.829	15.201	15.157	09.770
TiO_2	00.398	00.331	00.341	00.370
Al_2O_3	02.992	02.836	03.134	02.870
Fe_2O_3	01.552	01.335	01.617	01.380
MnO	00.037	00.035	00.032	00.010
MgO	01.178	01.074	01.003	00.880
CaO	42.925	42.430	41.550	45.730
Na_2O_3	00.118	04.005	05.694	00.010
K_2O	00.869	00.736	00.700	00.843
P_2O_5	00.234	00.217	00.239	00.186

T a b l e 3

XRD analysis: Summary of identified phases

Samples	Minerals	B1	B2	B3	B4
Whole rock	calcite	×	×	×	×
	quartz	×	×	×	×
Insoluble residue	quartz	×	×	×	×
	K-feldspar	×	×	×	×
	muscovite	×		×	×
	kaolinite				×
	goethite				×
Clay fraction	kaolinite	n.d.	×	n.d.	×
	illite	n.d.	×	n.d.	×
	smectite	n.d.	×	n.d.	×
	quartz	n.d.	×	n.d.	×
	K-feldspar	n.d.		n.d.	×
	Zeolite	n.d.	×	n.d.	

n.d.: not determined

filled with oxide mineral which is breaking down. Staining of the infill is intense around areas where such allochems are present.

The mineralogy of the clay fraction includes kaolinite, illite, smectite, quartz, and K-feldspar. Smectite and illite are structurally related to micas. Most of the interlayer water of smectites is lost on heating to 335 °C. Illite differs from muscovite in having less potassium and more silica. It may be formed during diagenesis by alteration of other clay species or be the result of post depositional weathering of muscovite and silicates. Kaolinite commonly results from weathering of feldspars and other silicates.

Although having visual resemblance, *sol ikħal* and limestone from the blue patches are lithologically different. Furthermore, they show notable geochemical (Fig. 3) and mineralogical differences. B4 represents a distinctive bed within the Lower Globigerina Limestone Member which Murray had described as dark bluish grey when freshly quarried and rapidly drying to pale grey when exposed [1, 3], a limestone which, similar to the blue patches, is soft and weathers poorly through successive flaking.

Although the matrix type common to all samples is foraminiferal, the petrographic characteristics of the *sol ikħal* is typical to the bed at the transition with the Lower Coralline Limestone Formation. The characteristic gradual rapid transition above the base of the Lower Globigerina Limestone Member from packstone to wackestone [2] indicates the horizon of the sample. Variations in the horizons are depositional and the result of diagenetic paleoenvironments [23]. The *sol ikħal* has been formed in deeper waters than the other beds within this Member. Phyllosilicates, mainly clays, decrease the degree of cementation and interparticle porosity. This accounts for its lower porosity when compared to the first and second quality limestone extracted from this Member, the latter referred to as *sol* [3, 24]. Oxidation accounts for the change in colour from dark blue to pale grey. The rate of oxidation is lower due to the phyllosilicates present. A similar context for the blue patches exists. On either side of the faults, a source of water ingress and seepage to the surrounding host rock, the blue patches will eventually turn into yellowish on further oxidation of the hydrous sodium carbonates. This supergene decomposition does not account for the significant presence of SiO_2, which may be attributed to quartz, the main non-carbonate industrial mineral present in this Member. This oxide is a diagnostic characteristic which distinguishes blue patches from *sol ikħal*.

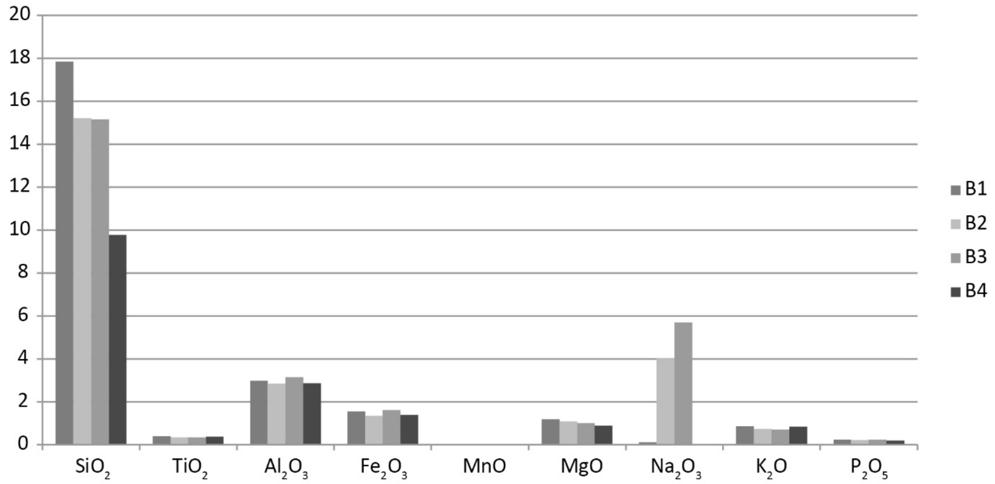


Fig. 3. Percentage distribution of the geochemical composition of the non-carbonate fraction

Conclusion. The new results corroborate and refine the findings documented in the seminal paper of Murray published over 125 years ago. The main non-carbonate mineral content of the blue patches and the *sol ikħal* which occur in the Lower Globigerina Limestone Member is quartz followed by K-feldspar and muscovite. Glauconite was detected in petrographical thin sections. Clay minerals are kaolinite, illite and smectite.

Although they both occur in the same lithostratigraphical unit, the *sol ikħal* has different composition compared to the blue patches. Despite being petrographically similar, the mineralogy and geochemistry are quantitatively different with minor qualitative variations. *Sol ikħal* has less insoluble residue content than the limestone from the blue patches. Also, it has higher calcium carbonate content and significantly less quartz. The SiO_2 content of the *sol ikħal* is $< 10\%$; in the case of the blue patches it is $> 10\%$.

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