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Mineralogical, Geochemical and Thermal Characterization of Precambrian Banded Marble, Gebel El Rokham, In Comparison with Serpentine-Related Carbonate, Eastern Desert, Egypt

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Abstract: ***The Precambrian marble is banded, composed of coarse translucent and fine grained bands. It is found as lensoidal mass intercalated with metsediments. XRD, DTA, XRF analyses and petrography reveal that, the composition of the banded marble is magnesite with minor phases of calcite, dolomite and hercynite. MgO and CaO of the coarse- and fine- grained bands are (41.8 and7.7 wt. %) and (43.6 and 3.8 wt. %) respectively. %.Fe_2O_3 (0.26 wt. %) and SiO_2 (0.87 wt. %) of fine-grained band are slightly higher than those of coarse-grained band. The serpentine-related carbonates are composed of white and translucent grains with massive texture. The fine-grains are composed of dolomite with minor phases of lizardite, vermiculite and anthophyllite. The translucent grains are composed of dolomite and minor phase of clinochlore. The serpentine–related carbonate has high concentrations of Cr, Ni, V, Co, Nb, Ta, La, Zr, Y and Sr. Abundances of Sr and Rb of translucent grains are higher than those of white grains of marble and serpentine–related carbonate which confirms their association with high content of calcium content in translucent grains of both types. The purity of the magnesite of the translucent band resulted in lowering the temperature of its decomposition (589 ºC) as compared with the fine-grained band (622 ºC).The low concentration of Cr, Ni, Y and occurrence of graphitic materials indicate para origin as a result of regional metamorphism of Mg-rich carbonate or magnesite of shallow marine environment.***

Keywords: marble, magensite, dolomite, XRD, DTA, XRF

1. Introduction

Marble is a metamorphic rock consisting of fine- to coarse-grained recrystallized calcite, magnesite and/or dolomite. It is often formed by metamorphism, may be either contact or regional metamorphic rock intermediate in grad between slate and mica schist. Marble deposits all over the world are mainly confined to metasedimentary belts. Pure marble (high calcium marble) is composed primarily of the minerals calcite or aragonite with total CaCO3content of between 97 – 99%, and pure dolomite is composed of 45.7% MgCO3 and 54.3% $CaCO_3$[1].Naturally occurring carbonate rocks are extremely important natural resources finding widespread applications and thus being placed among the important raw materials. Marble is a precious ornamental stone used by man very early in history. It could be said that marble quarrying and processing industries belong to the oldest industries of the world. The Ancient Egyptians, 5000 years ago, knew 40 different types of ornamental stones and worked chiefly with granite and some types of marble such as Alabaster. The naturally occurring carbonate of magnesium (Mg) is magnesite ($MgCO_3$) represent one of the natural sources for the production of magnesia (MgO). Magnesite deposit in British Columbia occurs within the foreland tectonostratigraphic belt and was deposited in a shallower marine. Metastable "precursor" (such as magnesium calcite) will change gradually into more and more of the stable phase (such as dolomite or magnesite) during periodical intervals of dissolution and re-precipitation [2]. [3] Explains the spary magnesite as sedimentary products in dolomitic facies. The genetic interpretation by sedimentary processes is being widely accepted. [4] explained the spary –type magnesite as have been formed in basins of epicontinental shelf, filled with pelitic sediments and magnesite deposits originated during further diagenetic phases .The Rubian magnesite deposit NW Spain is hosted within hundred meters thick carbonate/siliciclastic metasedimentary sequence that has been deformed and metamorphosed under low- to medium-grade conditions .The magnesite rock gradually becomes dolomite-rich; and passes to mica schist and slates [5] Today, marble is a differentiated product that is internationally traded. The price and value of marble depends on one hand on its natural characteristics (like quality, type and color) and on the other hand on technological processing (giving it a special shape, polish, size and thickness). Apart from chemical characteristics and composition, other basic physical properties of marble are also important in deciding its value such as water absorption, bulk density, compressive strength, modulus of rupture (bending strength) and durability are important features of the stone which is an important determinant of its price and value. Marble bands are of limited distribution and rhythmically alternated with bands of amphibole and mica schist forming a zone of about 20 m thick with sharp contacts against the surrounding rocks of Umm Nar area, Eastern Desert, Egypt [6].The metasediments are regionally metamorphosed to the green schist and amphibolite facies. The presence of coarse crystalline white marble bands interbanded with biotite gneisses in the southern part of the Eastern Desert, indicate without doubt their sedimentary origin. [7] Stressed that the low grade metasediments in the Eastern Desert are locally interbanded with very characteristic paramarble bands of 2 to 40 m thick, which may be accompanied by metamorphic iron ore bands as in

Wadi Kareim. Regarding the carbonates associated with the Egyptian serpentinite, the ultramafic rocks are largely converted to serpentinite and/or to mixtures of serpentine, talc, tremolite, chlorite, magnetite, and carbonates [8]. [9] found that the massive and sheared serpentinites of the Eastern Desert, Egypt have the same composition (antigorite, chrysotile, and lizardite, with minor carbonate, chromite, magnetite, magnesite, and chlorite).; [10] write that , magnesite occurs as sparse crystals or as veinlets. The origin of the carbonate alteration fluids remains to be elucidated, but [11 argued on the basis of C and Sr isotopic studies that pervasive carbonate alteration, affecting Egyptian ultramafic rocks, is a mixture of mantle derived and remobilized sedimentary carbonate. [12] Write that, these solutions were brought from serpentinized area following tectonic fractures. The magnesite precipitation is due to the decrease of partial pressure of supersaturated carbon dioxide hydrothermal fluids, most likely at temperature below 300 ºC.

The present work aimed to determine the mineralogical, geochemical and thermal characterization of the Precambrian marble as well as its origin by comparison with massive carbonates associated with the Egyptian serpentinite rocks

2. Methods of study

2.1 X-ray Diffractmeter analysis (XRD)

The mineral identification for banded marble and serpentine–related carbonate has been carried out by analysis of 4 samples (coarse- and fine-grained bands of banded marble and fine opaque -and translucent grains of the massive serpentine –related carbonate). The analysis are performed by A Philips X-Ray Diffraction equipment model X' Pert PRO with Monochromatic , Cu – radiation (λ = 1.54 Å) at 50 K. V. , 40 M.A. and scanning speed 0.02° / sec. were used at Central Laboratories Sector of the Egyptian Mineral Resources Authority , Ministry of Petroleum, Egypt. The reflection peaks between 2 Ø= 20 and 60, corresponding spacing (d Å) and relative intensities (I/I0) were obtained. The diffraction charts and relative intensities are obtained and compared with ICDD files. XRD patterns are shown in Figure 2.

2.2 Differential Thermal analysis (DTA)

The study banded marble is composed of two bands (white band and translucent band). The bands have been separated and each one was grounded with agate mortar and pestle and then sieved to less than 60 µm diameter. They are subjected to thermal analysis by (differential thermal analysis (DTA) at Housing and Building National Research Center), Cairo, Egypt. Each powdered sample was heated at heating rate 10 ºC / min. under static air atmosphere, from ambient temperature to 1000 ºC. DTA curves are shown in Figure 2.

2.3 X-ray fluorescence analysis (XRF)

The XRF analysis was carried out at Central Laboratories Sector of the Egyptian Mineral Resources Authority, Ministry of Petroleum, Cairo, Egypt. 4 samples are selected for major and trace elements analysis. (2 samples representing the coarse- grained band and fine-grained band of the marble and 2 samples representing the white and translucent grains of massive serpentine –related carbonate).The powder (< 74µm) samples are subjected to XRF analysis using X-Ray fluorescence equipment PW 2404 with six analyzing crystals. Crystals (LIF-200)., (LIF-220) were used for estimating Ca, Fe , K, Ti, Mn and other trace elements from Nickel to Uranium while crystal (TIAP) was used to determine Mg and Na . Crystal (Ge) was used for estimating P and crystal (PET) for determining Si and Al and PXL for determining sodium and magnesium. The concentration of the analyzed elements is determined by using software Super Q and Semi Q programs with accuracy 99.99% and confidence limit 96.7%.The estimation of the major and trace elements were done as powder pellets (Pellets method) which were prepared by pressing the powder of the sample in Aluminum Cup using Herzog presser and 10 ton pressure. On the other hand using fusion method in platinum crucible (Bead method) gives better results for light elements measurements, but owing to presence of phosphorus in phosphate and sulfur in sulphides in the sample which led to corrosion in platinum crucible, we used the pellet method as mentioned above. The data of XRF analysis is obtained in Table 1.

3. Previous work

[13] Presents geological and petrographical studies on Gebel El- Rokham marble, near Wadi El Miyah (Midway between Marsa Alam and Edfu~110 km from the Red Sea coast), Eastern Desert of Egypt (long, 33º 57' and latit. 25º 17'). It is outcropping as a huge ellipsoidal pocket. Its longest dimension measures about 80 meters in an east- west direction and its shortest dimension along a north south direction measuring about 40 meters (Fig. 1 a). Vertically it extends to about 40 meters and intruded by epidiorit. The marble is fine –grained, hard, white with saccharose appearance. It shows a banded structure (colored in part) and has pockets of calc –magnesian silicate minerals in contact with epidiorite masses. Its composition is calcitic with minor brucite and dolomite and hosted in slate, pelitic, psamopelitic metasediments and serpentine –related schist .It is originated from the dolomitic sediments .The serpentinite rocks are of two origins; one is derived from the ultrabasic rocks and not affected by gabbroic intrusion. The other is derived from the thermally metamorphosed dolomitic rocks (forsterite – diopside rocks) by the action of C_2O rich hydrothermal fluids. The present writer suggests that the unaffected serpentinite rocks may be represent a member of ophioltic rocks tectonically comes in contact with gabbroic rocks. For this reason, the marble is studied in comparison with the serpentine –related carbonate of Gebel EL –Rokham area.

Table 1: Major and trace elements analysis of marble and serpent-related carbonate

Samples	Serpentine- related carbonate		Banded Marble	
	White opaque grains	Translucent grains	Coarse-grained translucent	Fine-grained white
Major oxides (Wt. %)				
SiO_2	30.1	1.80	<0.01	0.87
TiO_2	0.02	0.01	0.01	0.01
Al_2O_3	<0.01	0.03	<0.01	<.01
Fe_2O_3	0.35	0.06	0.03	0.26
MnO	0.05	0.18	0.01	0.01
MgO	33.6	18.95	41.8	43.6
CaO	11.5	32.35	7.7	3.8
Na_2O	<0.01	<0.10	<0.01	<0.01
K_2O	<0.01	0.01	<0.01	<0.01
P_2O_5	<0.01	<0.01	0.01	<0.01
Cl	0.51	<0.01	<0.01	0.01
SO3	0.01	<0.01	<0.01	<0.01
L.O.I	23.7	46.18	50.31	51.13
Total	99.89	99.70	99.93	99.74
Trace elements (ppm) and their ratios				
V	9.1	9.4	5.4	<2
Cr	187	191.9	3.4	2.1
Ni	220	226.9	<2	<2
Cu	2.3	4.3	<2	<2
Zn	2.5	2.7	29.8	5.8
Co	9.1	10.5	<2	<2
Ga	4.7	10.6	10.1	9.7
Rb	5.8	8.8	7.3	6.3
Sr	840	939.3	476.1	389.2
Y	66.3	65.5	5.8	4.4
Zr	88.2	81.4	40.1	35.2
Nb	16.8	16.3	15.3	12.7
Ba	4.1	<2	<2	11.2
La	28	29	5.8	<2
Ta	2.1	2.2	<2	<2
Pb	2.8	2.6	<2	<2
Th	41	39	41.5	24.2
Nb/Zr	0.19	0.20	0.38	0.36
Zr/Y	1.33	1.24	6.91	8
Y/Nb	3.95	4.02	0.38	0.35
Mn/Mg	0.0019	0.012	0.0003	0.0003
Fe/Mg	0.012	0.065	0.0024	0.0069
Cr/Mg	0.0009	0.0017	1.35E-05	7.99E-06
K/Rb	10.97	7.23	8.72	10.10
Sr/Rb	144.83	106.74	65.22	61.78
Mg/Ca	82.78	15.42	156.71	144.60

4. Mineralogical and Thermal Characterization

Two types of carbonate,(obtained from the quarry of Gebel El Rokham).One in a banded structure known as marble interlayered with metasediments, graphite- ,talc- and mica -schist .The other type is massive carbonates associated with serpentinized ultrabasic rocks.

4.1 Banded marble

The rock is composed of two alternating bands; one is white (chalky appearance) with nodular structure (its density is3.782). The other band is coarse-grained and translucent (its density is 4.714). The marble rock not affected by the dilute hydrochloric acid .The density of the Egyptian Alabaster bands (calcitic) is 3.441 and 2.112 for coarse- and fine-grained bands respectively [14]. Each band is subjected to XRD and XRF analysis .The microscopic description and XRD analysis indicate that , the coarse-grained band is composed mainly of magnesite (Mg $_6C_6$ O_{18}) grains in a mosaic texture with minor phases of dolomite ($Ca_3Mg_3C_6O_{18}$) ,hercynite ($Fe_8Al_{116}O_{32}$) and calcite ($Ca_6C_6O_{18}$)(Fig 2 e .).The other band is white with muddy texture (Fig. 1 d)and composed mainly of magnesite ($Mg_6C_6O_{18}$), with minor phases of dolomite($Ca_{3.21}Mg_{2.79}C_6O_{18}$), and calcite ($Ca_6C_6O_{18}$) (Fig.2 f).Both white and translucent bands are subjected to differential thermal analysis (DTA) .The DTA curves (Fig.2 a) indicate two endothermic peaks for each band. The two peaks of coarse-grained band are at 588.9 and 806.66 ºC, corresponding to decarbonation of magnesite and calcite respectively. The two peaks of fine-grained band are at 621.88 and 813.19 ºC (slightly higher in temperatures than the associated coarse-grained band), corresponding to decarbonation of magnesite and calcite respectively.

The decarbonation temperatures of coarse- and fine-grained bands of the Egyptian Alabaster are 887 ºC and 875 ºC respectively for decarbonation of calcite. The white opaque band has another endothermic peak at 275 ºC corresponding to dehydroxilation of brucite and the coarse band has also another endothermal peak at 325 ºC corresponding to dehydration of physical water [14]. The absence of dehydration and dehydroxilation endothermic peak in the study banded marble suggest metamorphic origin not from carbonate rich solution as for Egyptian Alabaster. The microscopic description indicates that the banded marble have been affected by some degree of deformation emphasized from the presence of shearing planes, joints(now filled by microcrystals of carbonates) as well as presence of spindle clots of graphitic materials within the coarse-grained band (Fig.1 e).

It is interesting to write that, thermal analysis techniques give a clear picture of the constitution of magnesite mineral. The analysis can be used to find out the amount of calcite and dolomite (chief impurities of magnesite in a sample) [15].It may be seen further that the two samples of the study marble show two endothermic changes. The first one is the most intense and big peaks ataround 588.9 and 621.88 ºC for coarse- and fine-grained bands respectively represents the decomposition of magnesite evolving out C_2O. This points out; the main mineral of the study marble is magnesite. The other two peaks of coarse -and fine –grained bands are at around 806.66 and 813.19 ºC respectively are weak and small (Fig. 2 a).

Differential thermal curves for dolomite have been described by various authors although merely as a means for its identification in minerals. [16] found that the decarbonation curves of dolomite marble shows two endothermic peaks .the lower at 777.8 ºC corresponding to the decomposition of the dolomite structure accompanied by the formation of calcite and magnesium oxide. The higher temperature peak at 834 ºC represents the decarbonation of calcite with the evolution of carbon dioxide .The use of magnesium hydroxide (Mg $(OH)_2$) as a flame retardant and smoke suppressor is more environmentally friendly than the flame retardants based on antimony metals or halogenated compounds. Mg $(OH)_2$ can be produced by the hydration of magnesium oxide (MgO), which is usually produced industrially from the calcination of the mineral magnesite [15] and [17].

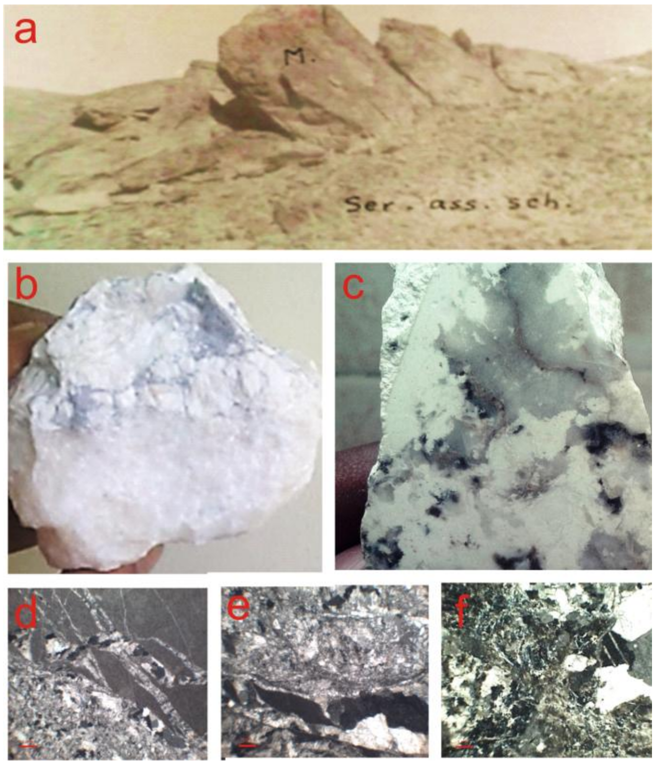

Figure 1: a) A photo showing lensoidal outcrop of marble, b) Banded marble, c) Massive serpentine-related carbonate, d) A photomicrograph showing coarse crystalline band in contact with very fine-grained band with muddy texture, e) A photomicrograph of coarse grained translucent band with spindle graphitic material, and f) A photomicrograph of massive serpentine-related carbonate showing coarse and fine grains of dolomite and fibrous serpentine minerals.

4.2 Serpentine- related carbonates

This sample is a fine –grained, dominated by, dolomite with minor vermiculite, lizardite, anthophyllite .Dolomite is distinguished from calcite by its cloudy appearance. Veinlets of carbonates crosscut the rock. Dolomite is present in two forms, one as coarse crystals, translucent with rhombic cleavage and twin lamellae. The other one is found as dusty grains and opaque. Serpentine minerals in contact with dolomite are pale and free of iron, therefore forming some grains of magnetite. The microscopic description and XRD analysis indicate that the massive serpentine –related carbonate rock is composed mainly of dolomite .The white and translucent grains are separated and subjected to X-ray fluorescence analysis (Table 1) and X-ray diffraction analysis (Figs. 2 b, c & d) .The translucent grains are composed mainly of dolomite ($Ca_3Mg_3C_6O_{18}$) with minor phase of clinochlore ($Mg_{4.5}Fe_{0.5}Al_{1.84}Si_{3.16}O_{18}$),while the white grains are composed of dolomite ($Ca_{2.99}Fe_{0.7}Mg_{2.31}C_6O_{18}$), lizardite ($Mg_3Si_2O_9$), vermiculite ($Mg_{12}O_{48}$ Si_{16}) and anthophyllite ($Mg_{28}Si_{32}O_{96}OH$).

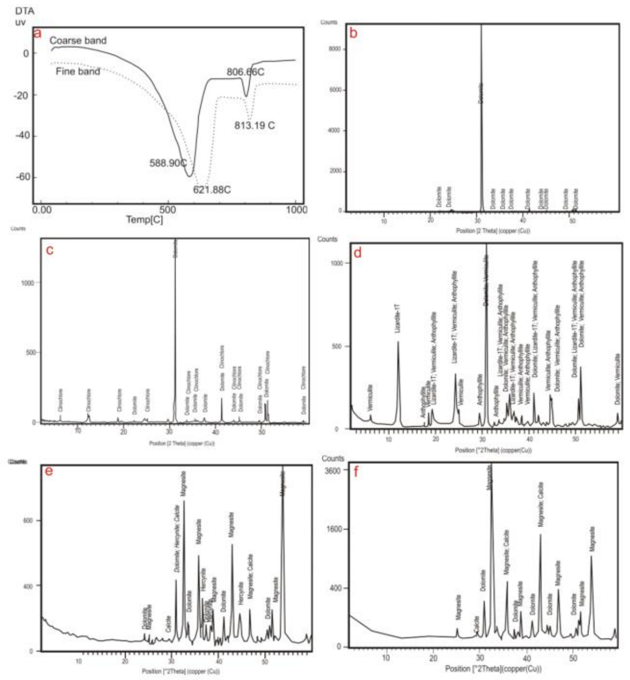

Figure 2: a) DTA curves of fine- and coarse-grained bands of marble, b) XRD pattern of translucent grains of serpentine-related carbonate, c) XRD pattern of white grains of serpentine-related carbonate, d) XRD pattern of massive serpentine-related carbonate, e) XRD pattern of coarse translucent band of marble f) XRD pattern of fine translucent band of marble.

5. Geochemical characterization

The composition of the banded marble is magnesite with minor phases of calcite, dolomite and hercynite. MgO and CaO of the coarse-grained band are 41.8 and7.7 wt. % and are equal to 43.6 and 3.8 wt. % for fine-grained band respectively .SiO_2, Al_2O_3,TiO_2 , P_2O_5, MnO, $K_2O$and Na_2O are less than 0.01wt % .Fe_2O_3 (0.26 wt. %) and SiO2 (0.87%) of fine-grained band are slightly higher than those of coarse-grained band. Sr of coarse band (476.1 ppm) is higher than fine band (389.2 ppm) which confirms its association with calcium content in coarse band. Cr, Ni, Co, V, Y, La, and Ta are very low .On the other hand; the serpentine-related carbonates are composed of white and translucent grains with massive texture. The white grains are very fine and composed of dolomite with minor phases of lizardite, vermiculite and anthophyllite. The translucent grains are composed of dolomite and minor phase of clinochlore .The serpentine –related carbonate is marked by high concentrations of Cr, Ni, V, Y and Sr as compared with the banded marble. The low concentration of Cr, Ni, Y and occurrence of graphitic materials in the study banded marble indicate Para origin. The element ratios Mn/Mg, Cr/Mg, Fe/Mg, Ca/Mg, Sr/Rb, Y/Nb, Nb/Zr of dolomite carbonate after serpentine are higher than those of magnesitic marble except Zr/Y is lower. K/Rb of white opaque grain of marble and dolomite after serpentine is higher than that of translucent grains of both types. The mentioned element ratios of dolomitic carbonates (opaque and translucent grains) are more or less similar to each other. As well as these ratios of the opaque and translucent bands of the study banded marble are comparable .The Cr and Ni enrichment in dolomite is due to mixture of other trace mineral phases derived from parent ultramafic rocks. The dolomite is considered to be the result of direct replacement of serpentine by CaO and CO_2-bearing solutions .On account of ionic radius of iron was unable to enter the dolomite lattice and therefore cast out, some forming magnetite grains. In addition to this role of iron, however, serpentine included in and bordering the dolomite is usually paler in color, suggesting leaching of iron by the dolomitizing solutions and, most ankerite-poor dolomites was assigning to a metasomatic origin. Stern and Gwinn (1990) found that the K and Rb for the sedimentary carbonates are 0.05, 13 ppm and for the intrusive carbonates are 0.34 ppm and 106 ppm. Sr has a wide range of concentrations from 76 to 1266 ppm of sedimentary carbonates while the intrusive carbonate has Sr concentration vary widely from 35 to 1676 ppm with a mean of 383ppm.The present marble has lower contents of K_2O(< 0.01), Rb (6.3 and 7.3) , Sr (389.2 and 476.1), so it is derived from sedimentary carbonates. Some of the banding is due to differences in chemical composition (silica, magnesium) between original carbonate sediments beds that would not have been obvious in the original sediment .The recrystallization in solid state under the action of heat and pressure play an important role in differentiation. The presence of graphitic materials inhibit the diffusion rate of atoms to grow so, the carbonate layers containing graphitic material exhibits fine-grained texture [18].The translucent band of marble and the translucent grains of serpentine –related carbonate characterized by higher contents of Sr and Rb than the associated opaque and white grains. This means that Sr and Rb are high in pure or translucent mineral. These highest contents confirm their association with high calcium content in the translucent grains [1].

6. Results and Discussion

Banded marble of Gebel El Rokham outcropping as a lensoidal mass hosted in metasediments and intruded by gabbroic and granitic magmas. The marble rocks are white, hard, heavy, and exhibits banded structure. One band is very fine-grained, white, and opaque with some graphitic materials. The other band is translucent and coarse-grained. On the other hand, the serpentinite rocks occur as lensoidal masses hosted within the metasediments .Sometimes, the serpentine have been altered to massive carbonate rocks and talc schist rocks. The carbonate rocks are hard, white and composed of chalky white opaque grains and translucent grains. The marble is composed mainly of magnesite with minor phases of calcite while serpentine-related carbonate is composed mainly of dolomite with minor phases of lizardite , vermiculite and anthophyllite .The geological setting of the study marble as well as its mineral composition together with negligible concentration of Cr , Ni , V, Mn, Fe and Y suggest without doubt a sedimentary origin. [19] Has shown that the dolomitic marble derived from ultrabasic rocks has high concentration oF Cr, Ni, Co and Sc but the marble derived from sedimentary carbonate by metamorphism or hydrothermal solutions has low or negligible contents of these elements. The magnesite was deposited on shallow marine platform environment and the calcitic marble, deposited on open-marine environment, while dolomitic marbles, deposited on less open marine environment. This points to a process of chemical differentiation from open sea to landwards, more calcic in the first to more magnesian in the confined environment, in which waters are progressively purified from Ca through preferential precipitation of calcic carbonates, which increases the Mg/Ca ratio, and lead carbonate deposits towards the magnesite ([20].The carbonate sediments and their host slaty , pelitic and psamopelitic have been regionally metamorphosed up to low -to medium grade . The magmatic intrusion imparts thermal metamorphism along the contacts giving hornfelsic rocks. Marble rocks in contact with the gabbroic mass have been invaded and partly replaced by the silica –rich solution resulted in producing some colored bands and pocket of calc-magnesian silicate minerals [13]. [20] have been studied five important magnesite mines form a sequence of lenses that extend, discontinuously and found that, magnesite deposits pass gradually into metadolomites and then to almost pure calcitic marbles. This group is hosted in green schist to amphibolite-facies metavolcanic-sedimentary sequence crosscut by basic and granitic intrusions of variable size. More than 70 magnesite occurrences have been reported in British Columbia. Most of the significant deposits are hosted by sedimentary rocks that were deposited in shallow marine environments. Most of the promising sparry magnesite deposits are hosted by sedimentary rocks of Precambrian to Cambrian age in southeastern British Columbia [21].

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