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Geochemical and petrogenetic characteristics of the marble deposit in Itobe area, Kogi state, Central Nigeria

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ABSTRACT

Geochemical investigations of the marble deposit in Itobe, Kogi state, central Nigeria shows that the two outcropping masses (mass I and mass II) of the marble deposit are calcitic in composition. The two masses of the marble deposit may have been deposited in shallow structural basins, and mass I with a much higher MgO content (10.4%) was deposited in a much deeper basin than mass II with a much lower MgO content (0.74%). The Mg in the marble must have been co-precipitated with the Ca from hyper-saline waters under anoxic conditions with microfauna and flora playing a vital role. The Itobe marble, as with other metamorphic carbonate rocks, occur commonly within continental environments along which crustal distension has occurred. The association of the marble deposit with quartzite, quartz schist, gneissic granite, pegmatite and quartz veins suggests deposition in a rift environment of rapid subsidence with contemparenous magmatism in the basins formed, followed by closure which led to the deformation of the sediments.

Keywords: Marble, Mass, Major, Carbonate and Content

INTRODUCTION

Marble, a major raw material for industries, results from the metamorphism of limestone, a carbonate sedimentary rock formed at the bottom of lakes and seas as silt and organic matter settle from the water body to the bottom. For practical purposes, pure marble (high calcium marble) is composed primarily of the minerals calcite or aragonite with total CaCO₃ content of between 97 – 99%, and pure dolomite is composed of 45.7% MgCO₃ and 54.3% CaCO₃ or 30.4% lime (CaO) and 21.8% Magnesia (MgO), [Boynton]. Calcite and dolomite are the major constituents of marble and often coexist in chemical equilibrium.

The Itobe marble body is located about 1km from Itobe town along the Ajaokuta-Anyigba road, Kogi state, central Nigeria. The study area lies between longitudes $6^{0}40'$ E and $6^{0}48'$ E and latitudes $7^{0}22'$ N and $7^{0}30'$ N (Fig.1).



Fig.1: Geological sketch Map of Nigeria Showing the Location of the Study Area (After Obaje, 2009)

[Hockey *et al.*,] in the regional study of the geology of the Lokoja – Auchi area described the Itobe marble as a dark grey, fine- medium grained narrow band of marble.

[Akoh] based on limited geochemical data described the Itobe marble body as a dolomitic marble. This study seeks to determine the geochemical characteristics of the Itobe marble and the possible origin of the marble.

Field Geologic Occurrence

The Itobe marble deposit is associated with crystalline rocks of the Precambrian basement complex including biotite schist, mica (muscovite & biotite) schist, quartz- muscovite schist, quartzite, biotite- hornblende schist, quartz schist, gneissic granite and minor intrusive rocks including pegmatite, quartz and quartzo-feldspathic veins. The marble body occurs within a host rock of mica/ quartz schist and feldspathic quartzite (Fig. 2). Two outcrops (described as mass I and mass II) of the marble have been identified in the study area. Mass I with a dimension of 1.4m x 0.6m, trends NE - SW with a strike azimuth of 033⁰. It is poorly exposed at the road cut about 150m to Alo I village along the Anyigba – Itobe road. It is light grey in colour and fine grained in texture. Mass II, which is about 800m from mass I on a NE- SW axis, is a much larger marble body and outcrops on the Ayanka hill as massive boulders. It is medium grained in texture, dark grey in color and rises from an elevation of 132m to 138m on the hill. It has a sharp contact with the quartz schist below and the feldspathic quartzite overlying it.



Fig. 2 Geologic Map of the Itobe Marble Area (After Onimisi, 2013)

MATERIALS AND METHODS

X- Ray Fluorescence (XRF) Spectrometric analysis of whole rock samples to determine the major elements composition of the marble deposit was carried out. Major element oxides determined include SiO₂, CaO, MgO, Al₂O₃, Na₂O, K₂O, SO₃ and total Fe as Fe₂O₃. Loss on ignition (L.O.I) of the marble samples was determined gravimetrically. Forty (40) samples of mass II and ten (10) samples of mass I of the Itobe marble were subjected to X-Ray fluorescence (XRF) spectrometry.

Procedure for XRF Spectrometry

The marble samples for the XRF analysis were prepared by subjecting a wet portion of the rock samples (weighing about 5kg) to wet grinding on a silicon carbide disk mill to remove saw mark and unwanted materials. The samples were ultrasonically washed in distilled water and then methanol for 10 minutes and dried at 110 $^{\circ}$ C for at least 2 hours. Larger pieces were reduced to < 1cm in diameter in a Spex shatter box using a tungsten carbide grinding

vessel for 1 - 2 minutes depending on the size of the sample. The powder was transferred to a clean paper and then to a sample vial and labeled.

Borate glass discs were prepared for the X-ray analysis by mixing 2g of ignited sample powder and 4g lithium tetraborate ($Li_2B_4O_7$). The mixture in a platinum crucible, were melted together in an induction melting oven for some minutes. The melt obtained was turned into a disc mould and allowed to consolidate into a glass disc. Whole rock analysis for major elements Al, Na, K, Ca, Mg, total Fe (Fe²⁺, Fe³⁺), and S were made on the lithium borate discs using an ORTEC 6111 energy dispersive X-Ray Fluorescence spectrometer. A 30 Mci, 241 Am. Isotropic X-ray sources served as an excitation source. The spectral were collected for 30 minutes each and evaluated using AXIL computer program.

Instrument settings were at count of 800-1000 for standards and 100 seconds for major elements. The accuracy of analysis ranges from ± 1.0 to $\pm 3\%$.

RESULTS AND DISCUSSION

The mean and ranges of the major element oxides, trace elements and the calculated values of CaCO₃, MgCO3, determined from the XRF analysis of the Itobe marble are presented in table 1.

Table 1: The Mean and Ranges of the Wt % of the Major Element Oxides, the Calculated Values of CaCO₃, MgCO₃, and the Trace Element content of the Itobe Marbles

	Mass I		Mass II			
Major element Oxides (%)	Mean N = 10	Range	Mean N = 40	Range		
SiO ₂	3.78	6.13 – 2.47	6.39	14.87 - 1.60		
Al ₂ O ₃	0.27	0.48 - 0.19	0.52	1.61 - 0.10		
Fe ₂ O ₃	0.58	1.40 - 0.16	0.44	0.96 - 0.14		
MgO	10.4	22.54 - 0.23	0.74	3.39 - 0.09		
CaO	45.9	52.47 - 39.16	50.14	53.69 - 44.41		
K ₂ O	0.06	0.06 - 0.04	0.13	0.39 - 0.03		
Na ₂ O	0.06	0.11 - 0.10	0.03	0.25 - 0.01		
SO ₃	0.02	0.04 - 0.01	0.07	0.55 - 0.11		
CaCO ₃	81.61	93.40 - 69.71	89.26	95.57 – 79.05		
MgCO ₃	21.20	47.11 - 0.48	1.38	7.09 - 0.19		
L.O.I	38.15	42.89 - 31.70	41.27	43.39 - 38.75		
Trace element (ppm)	Mass I		1	Mass II		
	Mean N=7	Range	Mean N=9	Range		
Ni	13	15-9	14	17 -12		
Cr	11	13-9	12	17-9		
Zn	19	20-15	32	45 - 27		
Sr	34	38-22	41	60 - 17		
Mn	24	24-20	118	359 - 30		

Major Element Geochemistry CaO/MgO

In most carbonates of metamorphic terrain, CaO and MgO are the most important elemental oxides. In calcitic type marble, CaO is usually in the order 50 - 54% while MgO is < 15%. Dolomitic or Magnesian type marble on the other hand have CaO values generally in the range of 28-31% and MgO values in the range of 15-21% [GoldSchmidt *et. al*,]. A value of MgO greater than 1% in limestone (the parent rock from which marble is formed) suggests that the mineral dolomite is present [Brownflow]. The Ca⁺⁺/Mg⁺⁺ ratio has been generally accepted as a classification criteria for carbonates; those with Ca⁺⁺/Mg⁺⁺ of the order of 1.0 -1.5 are dolomitic while those of the order of 8 – 10 are calcitic [Talbor].

The marble samples from mass I have a relatively high MgO concentration (>10%) compared to that of Mass II with less than 1% MgO (Table 1). The MgO content in the two marble bodies range from 0.23% to 22.54% with a mean value of 10.40% in mass I, and 0.09% - 3.39% with a mean value of 0.74% in mass II.

Mass II has a higher CaO content with CaO values ranging from 44.41% - 53.69% with a mean value of 50.14%. CaO content in mass I range from 39.16% - 52.47% with a mean value of 45.9%.

The average ratio of CaO to MgO in masses I and II is 4:1 and 67:1 respectively. The mean CaO and MgO content of 50.14% and 0.74% respectively and Ca/Mg ratio of 67:1 for the marble samples of mass II are comparable to those of typical calcitic marbles (Table 2). Mass I with a mean CaO and MgO content of 45.9% and 10.4% respectively and a Ca/Mg ratio of 4:1 although comparable with typical calcitic marbles (Table 2), has a relatively high MgO content. Thus the the two masses of Itobe marble are calcitic marble containing dolomite crystals in varying proportion.

Table 2: A Comparison of the Chemical Composition of Itobe Marble Body with Typical Calcitic and Dolomitic Marbles

	Group A Typical Calcitic Marbles				Grou Itobe M (This s	ıp B /Iarble study)	Group C Typical Dolomitic Marbles			
Major Element Oxides	1	2	3	4	5	Mass II N= 40	Mass I N=10	6	7	8
SiO ₂	0.71	0.90	1.18	0.25	11.92	6.39	3.78	0.49	2.40	1.98
TiO ₂	n.d.	0.02		n.d	0.13			n.d.	0.01	0.01
Al_2O_3	0.52	0.07	0.08	0.35	2.39	0.52	0.27	0.02	0.92	0.13
$(Fe_2O_3)_T$	0.34	0.18	0.07	-	1.04	0.44	0.58	0.06	0.04	0.36
MnO			0.03	-	0.03			0.03	0.009	0.03
MgO	0.35	0.42	1.75	0.15	0.67	0.74	10.4	20.70	19.60	20.84
CaO	54.75	54.01	53.64	56.12	45.00	50.14	45.9	28.94	31.82	31.04
Na ₂ O	0.01	0.02	0.01	0.01	0.39	0.03	0.06	0.01	0.05	n.d.
K ₂ O	0.01	0.08	0.02	0.03	0.70	0.13	0.06	0.01	0.007	0.07
P_2O_5		0.02			n.d.				0.045	n.d.
CaCO ₃	97.71		95.72			89.26	81.61	51.69		
MgCO ₃	0.78		3.67			1.38	21.20	43.34	44.09	
L.O.I	42.01	43.72		-		41.27	38.15			

1. Shapfell marble (Calcitic), [Dowrie et *al*]

2. Jakura marble (Calcitic), [Okunlola]

3. Ososo marble (Calcite), [Emofurieta et al]

- 4. Mfamosing limestone [Dada]
- 5. Zambezi belt marble (Calcitic), [Munyanyiwa and Hanson]
- 6. Igbeti marble (Dolomitic), [Emofurieta et al]

7. FCT, Abuja marble (Dolomitic), [Davou and Ashano]

8. Zambezi belt marble (Dolomitic), [Munyanyiwa and Hanson].

Percentage of CaCO₃ and MgCO₃

Pure carbonates are believed to have a total carbonate content $(CaCO_3 \text{ or } CaMg(CO_3)_2 \text{ of } 70\%$ and above while impure carbonates have between 40 - 70%. Other oxides like SiO₂, TiO₂, MnO, and Na₂O are usually less than 1%, and are regarded as constituting impurities [Lippman].

The average $CaCO_3$ for masses I and II are 81.61% and 89.26% respectively. The average MgCO₃ content calculated for masses I and II are 21.80% and 1.38% respectively. A plot of the % weight of CaCO₃, MgCO₃ and others on a triangular diagram of classification of carbonate rocks adopted from [Carr and Ronney], (Fig. 3) shows the two marble bodies fall within two subdivisions of the carbonate rocks. Mass I with average CaCO₃ content of 81.61% and MgCO₃ content of 21.80% falls within the impure Calcite-Dolomite (I) field while mass II with average CaCO₃ content of 1.38% falls within the impure calcite marble (C) field. The host rocks of the marble body with less than 50% of total carbonates are non carbonate rocks.



Fig. 3: Classification of the Itobe Marble [After Carr & Rooney]

Silica Content

Silica in carbonate rocks is contributed from both silicate minerals and chert nodules resulting from the influx of near shore materials into the basin of deposition of the original limestone body prior to metamorphism [Brownflow]. The silica content in the two masses of the marbles samples vary widely. It ranges from 2.47% to 6.13% (average value = 3.78%) in mass I, and from 1.60% to 14.87% (average value = 6.39%) in mass II (Table 1). These values are comparable to those of similar carbonate bodies elsewhere (Table 2). The relatively higher content of silica in mass II compared to mass I can be attributed to the relatively shallower depth of deposition of the pre- metamorphic limestone body in mass II relative to mass I.

Alkali (Na₂O + K₂O) Content

The alkali elements such as Na and K which are indicative of salinity levels have been very useful in interpreting depositional and lithification conditions prior to metamorphism of carbonates [Land and Hopp].

The concentration of the total alkalis (Na_2O+K_2O) in the two marble bodies in the Itobe area is very low, generally less than 1%. This value is similar to the alkali content of typical marble bodies (Table 2)

According to Clarke (1924), Na and K concentration in marbles tend to decrease with increase in salinity. The low values of total alkali content in the Itobe marbles indicates that the environment of deposition of the original carbonate materials that were metamorphosed to marbles in the Itobe area must have been a shallow, highly saline environment, and mass II with a higher total alkali content of 0.16% must have been formed in the shallower part of the basin than mass I with a lower total alkali content of 0.12%.

Other oxides (Al₂O₃ and Fe₂O₃)

The other oxides $(Al_2O_3 \text{ and } Fe_2O_3)$, are generally low, less than 1%, and may be indicative of the absence of aluminosilicates in the marble

Loss on Ignition (L.O.I)

L.O.I reflects the content of volatiles (CO₂, H_2O) present in the marble bodies. The average values of L.O.I of the marble bodies in the study area are 38.95% and 41.27% for masses I and II respectively. In general, L.O.I values are high. This indicates high volatile content and by implication high carbonate content since it is synonymous with the

evolvement of carbon dioxide after heating at 900 °C [Olatunji]. Mass II with a higher L.O.I. value of 41.27% has higher carbonate content than mass I with L.O.I value of 38.95%.

A comparison of the mean composition of the Itobe marble bodies with average values for unmetamorphosed carbonate rocks (Table 3) shows that the Itobe marbles are similar to unmetamorphosed impure limestone.

Major Oxides (Wt %)	Dolostones (Pettijohn, 1975) (n=3)	Relatively pure limestone	Impure limestone	Itobe marbles (This study)		
		(retujoni, 1975) (n=3)	(n= 2)	Mass I N = 10	Mass II N = 40	
SiO ₂	3.98	1.88	10.61	3.78	6.39	
TiO ₂	0.04	0.01	0.07	N.d.	N.d.	
Al_2O_3	0.78	0.83	4.30	0.27	0.52	
*Fe ₂ O ₃	0.64	0.26	2.97	0.58	0.44	
MnO	0.02	0.01	0.22	N.d.	N.d.	
MgO	20.26	2.75	2.01	10.40	0.74	
CaO	29.09	50.89	41.90	45.90	50.14	
Na ₂ O	0.14	0.06	1.38	0.06	0.03	
K ₂ O	0.04	0.01	0.56	0.06	0.13	
S	0.06	0.18	0.13	0.02	0.07	
P_2O_5	0.32	0.01	0.21	N.d.	N.d.	

Table 3: Mean Compositions of Itobe Marbles Compared to Compositions of Unmetamorphosed Carbonate Rocks

* Fe_2O_3 is measured as total Fe

Trace Element Geochemistry

Since some sedimentary rocks have unique assemblages of trace elements, a study of these elements in metamorphic rocks provides a unique way to guess the nature of the pre-metamorphic material. Immobile trace elements such as the Rare-Earth Elements and the High Field Strength elements are important for provenance determination of pelitic rocks [Taylor and McLennan] because their concentrations often reflect those of their source rock. Most of the rarer elements are more abundant in shale than in sandstones and limestone [Konrad et al]. The major exceptions are strontium and manganese, which are markedly enriched in carbonate sediments. The enrichment of strontium in limestone is accounted for by the fact that Sr^{2+} substitutes readily for the very similar ion Ca^{2+} . The smaller but appreciable concentration of manganese in carbonates is probably also attributable to similarity in ionic size.

A comparison of the trace elements (Ni, Cr, Zn, Sr and Mn) content in the Itobe marbles with that in sedimentary carbonates (the protolith of marbles), is shown in Table 4.

Table 4: A Comparison of Some Trace Elements of Itobe Marbles and that of Sedimentary Carbonate Rocks

Trace		Itobe marbles (This study)				Carbonate rocks (Turekian and Wedepohl, 1961)		
(ppm)	Ma	Mass I Mass II			Challers Deer			
	Mean N=7	Range	Mean N=9	Range	Shallow Sea	Deep Sea		
Ni	13	15-9	14	17 -12	20	30		
Cr	11	13-9	12	17-9	11	11		
Zn	19	20-15	32	45 - 27	20	35		
Sr	34	38-22	41	60 – 17	610	2000		
Mn	24	24-20	118	359 - 30	1100	1000		

The Sr content in Itobe marbles range from 22 to 38 with an average value of 33 in mass I, and from 17 to 60 with an average value of 41 in mass II. The Sr content in the Itobe marbles is much lower than that in sedimentary carbonates. Mn content in Itobe marbles range from 20 to 24 in mass I with a mean value of 22. In mass II the Mn content ranges from 30 to 359 with a mean value of 127. The Mn content in Itobe marbles is much lower than that in sedimentary carbonates. The low content of Sr and Mn in the Itobe marbles relative to that in sedimentary carbonates may be attributed to the substitution of Sr and Mn by Ca during the recrystallization of the mineral grains at higher temperature.

The concentration of the immobile elements (Ni and Cr) in the Itobe marbles is relatively low and very similar to that in sedimentary carbonates (Table 4). The concentrations of Ni and Cr in the Itobe marble bodies therefore reflect those of their source rocks.

Variation Diagrams

A series of major element oxide plots for the Itobe marble is presented in Figs. 4 - 13. Plots of CaO, MgO, (FeO)_T, Na₂O+K₂O vs. SiO₂ were carried out for the two masses of the Itobe marble. SiO₂ was used as abscissa in these plots because it shows substantial variations among the marbles bodies. The variation plots for the two outcrops of the marble show a similar trend except the plots of Na₂O+K₂O vs. SiO₂ which show opposite trend.

CaO, MgO and FeO (Total) exhibit a well defined negative correlation with SiO_2 and this reflects the introduction of chert during the deposition of the parent limestone of the marble body. The variation plot of CaO vs MgO (Figs. 12 & 13) shows an inverse relationship. This is reflected in the inverse relationship in the proportion of calcite to dolomite in the marble.

The opposite trend of the plots of $Na_2O + K_2O$ vs. SiO_2 for masses I and II shows a variation in the salinity level between the two masses.



Fig 4: Plot of Wt% of Na₂O+K₂O vs. SiO₂ for Mass II



Fig. 5: Plot of Wt% of Na₂O+K₂O vs. SiO₂ for Mass I







Fig. 9: Plot of Wt% of FeO (Total) vs. SiO₂ for Mass II







Fig. 11: Plot of Wt % of MgO vs. SiO2 for Mass II



Nature of Protolith

To understand the pre-metamorphic parent material of the Itobe marbles, a discriminant plot of Na_2O/Al_2O_3 vs. K_2O/Al_2O_3 of [Garrels and Mckenzie] done for the two marble bodies in the study area (Figs. 14 and 15) shows that masses I and II of the Itobe marble fall within the field of sedimentary and metasedimentary rocks. This conforms to the observation from field evidence which shows the crystallinity of the marble and the interbedding of the marble with schist and quartzite, a characteristic used in classifying them as metasedimentary in origin.



Fig. 14: Na₂O/Al₂O₃ Vs K₂O/Al₂O₃ Variation Diagram of Itobe Marble-Mass I (After Garrels and Mackenzie, 1971)



Fig. 15: Na₂O/Al₂O₃ Variation Diagram of Itobe Marble- Mass II (After Garrels and Mackenzie, 1971)

DISCUSSION AND CONCLUSION

The major element composition of the Itobe marble shows it is similar in composition to calcitic type marble (Table 2) and relatively pure limestone (Table 3). The similarity between the two masses of the marble is apparent from the similarity in trend in the geochemical plots of the major element oxides (Figs. 4 - 13). Most of the major element

oxides show a negative correlation relative to SiO_2 . CaO shows a well defined negative correlation with SiO_2 and this probably reflects the admixture of the carbonates with chert. The opposite trend of the plots of $Na_2O + K_2O$ vs. SiO_2 for masses I and II shows a variation in the salinity level between the two masses.

The origin of dolomites in marble has been a controversial issue - "the dolomite problem". McKenzie (1991) asserted that dolomite, despite its thermodynamic stability and abundance in ancient rock records, are rarely found in Holocene environments. [Oates] investigated the limestone deposits of Tangayinka territory and noted a gradual decrease in dolomite from the Precambrian marble to its total absence in recent limestone. The absence of dolomite in recently formed limestone has led to a general conclusion that the presence of dolomite in marbles is by replacement of Ca by Mg during metamorphism. The discovery of dolomite in recently formed carbonates has added a new dimension to this controversy. [Vasconceles and McKenzie] discovered a large modern-dolomite Formation in Lagoa, Vernella, a shallow water, isolated coastal lagoon, east of Rio de Janerio, Brazil, and concluded that under anoxic conditions, Mg co-precipitates with Ca. [Davou and Ashano] opined that the homogeneity of the FCT, Abuja, Nigeria , dolomite marbles as revealed by petrographic studies is an indication that dolomite is probably one of the initial mineral constituents of the limestone, and that the dolomite was probably of precipitatory origin rather than replacement. This may also be true of the dolomitic crystals present in the Itobe marble as the marble is essentially a calcitic marble.

Thus the protolith of the Itobe marble is a relatively pure limestone in which dolomite is present in varying proportions.

The limestone protoliths of the Itobe marble deposit were deposited within shallow narrow basins which were probably initiated in structurally controlled depressions. Mass I with a higher MgO content must have been deposited in a deeper basin than Mass II as revealed by the lower percentage of silica impurities in Mass I than Mass II. The associated MgO in the marble must have been co-precipitated with the Ca from hyper-saline waters under anoxic conditions with micro-fauna and flora playing a vital role.

The Itobe marble occurs in association with Precambrian basement rocks comprising biotite schist, mica schist, quartz-schist, biotite hornblende schist, felsdapthic quartzite, quartzitic schist, gneissic granite and minor intrusive rocks including pegmatite and quartz veins. Plots of the Itobe marble samples on the Na₂O/Al₂O₃ vs. K₂O/Al₂O₃ curve of [Garrels and McKenzie]shows that the Itobe marble body falls within the field of sedimentary/metasedimentary field, thus confirming field and petrographic evidence which shows the interbeddding of the marble with schists and quartzites, a characteristic used in classifying them as metasedimentary in origin. Metamorphic carbonate regions, as in the study area, occur commonly within continental environments along which crustal distension has occurred [Elueze; Hoefs]. The occurrence of shallow water epiclastic carbonate bodies in association with deformed quartzite, quartz schist, gneissic granite, pegmatite and quartz veins suggests deposition in a rift environment of rapid subsidence with contemparenous magmatism in the basins formed, followed by closure which led to the deformation of the sediments.

The evolution of the rocks in the study area started with a period of orogenic deformation and metamorphism probably in the Kibaran (1200-700Ma) which resulted in the weak deformation and metamorphism of earlier deposited sediments in the rift basin. This actually resulted in the formation of marble, quartzite, quartz schist and rejuvenation of the basement at depth.

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