

RESEARCH ARTICLE

GEOCHEMICAL ASSESSMENT OF LIMESTONE DEPOSITS AROUND OMI ALAYO - OKELUSE, SOUTHWESTERN NIGERIA

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ABSTRACT

Limestone deposits exposed at Omi-Alayo and Okeluse Forest Reserve were investigated in this study. 10 representative samples collected from both locations and were subjected to geochemical analyses using X-Ray Fluorescence method. Major and trace elements were employed for the determination of depositional environment. The results for the major oxide contents showed that mean values of CaO content ranges from 48.10 – 48.89% with mean value of 7.23% for SiO₂, Fe₂O₃, MgO and Al₂O₃ concentration have mean values of 1.56%, 0.76% and 1.92% respectively. Other oxides TiO₂, Na₂O, K₂O and P₂O₅ are low in concentration. For the loss of ignition, it varied from 35.24 to 41.07 with an average value of 38.69% indicating high carbonate content for the limestone. Limestone classification of Ca/Mg ratio and reciprocal MgO/CaO showed ratios range of 54.05 – 86.46 and 0.011-0.024 respectively. High content of Sr recorded for the limestone samples reveals a saline environmental condition for the deposits. Overall results showed that the Omi-Alayo and Okeluse limestone are pure limestone and were deposited in a shallow marine environmental condition and suitable for cement production.

KEYWORDS

carbonate, strontium, geochemical, cement, Omi-Alayo-Okeluse

1. INTRODUCTION

Limestone is an important ingredient in the production of cement worldwide. So and many by-products has been produced from limestone. Examples are in ceramics, pottery, paints manufacture, refractory, and in fertilizers. However, detailed geochemical analyses are required for it to be considered suitable for the production of cement. Few published works can be retrieved from literature on the limestone deposits exposed at the Okeluse area of southwestern Nigeria. Several workers had reported occurrence of limestone deposits in Nigeria (Ola, 1977; Gwosdz, 1996; Akinmosun et al., 2005, Ehinola et al., 2012). Works of a group researchers was on depositional environment and geophysical mapping/reserve estimation of Arimogija - Okeluse limestone while that of Edema et al (2016) was on evaluation of limestone of Aimogija-Okeluse limestone, S.W Nigeria. Detailed geochemical analyses on these deposits in order to assess their suitability for cement production necessitated this present study. Therefore, intends to combine both major elemental oxides (wt.%) and trace element geochemistry to decipher the quality of the limestone at both locations as well as predict the condition of formation of the limestone deposits under investigation. The study areas, Omi-Alayo - Okeluse Forest Reserve is located in the Eastern flank of the Dahomey basin in southwestern Nigeria. It lies within Latitudes 6°. 45' N- 6°. 47' N and Longitudes 5°. 33' E- 5°. 37' E (Figure 1). The area is easily accessible with major roads and numerous footbapths abound in the area. Drainage pattern is dendritic in nature (Figure 2). Elevation ranges from 150m to 300m above the sea level (Figure 2). The relief is high and major rivers dissect the hills along the valley. The climate is typical rain forest

characterized by thick forests. Two seasons are recognized (Iloje, 1976). Rainy season prevails between March and October while the dry harmattan season commences in November and ends in March. The humidity is high and average mean temperature is 27°C.

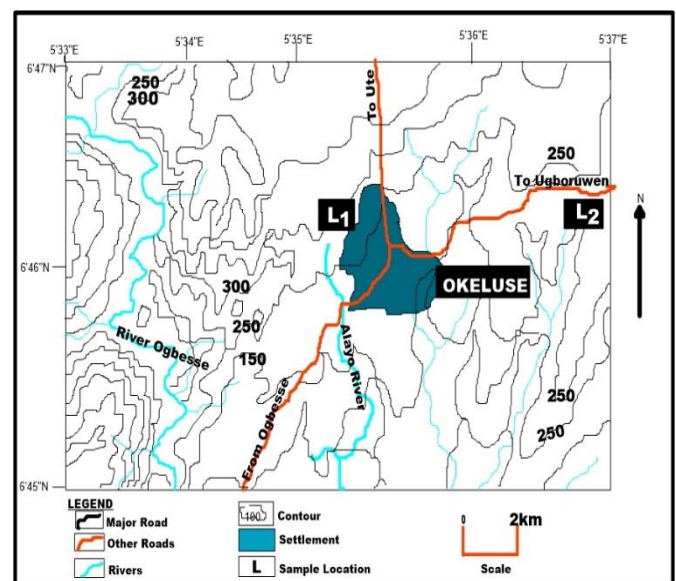


Figure 1: Location map of the study area, Okeluse

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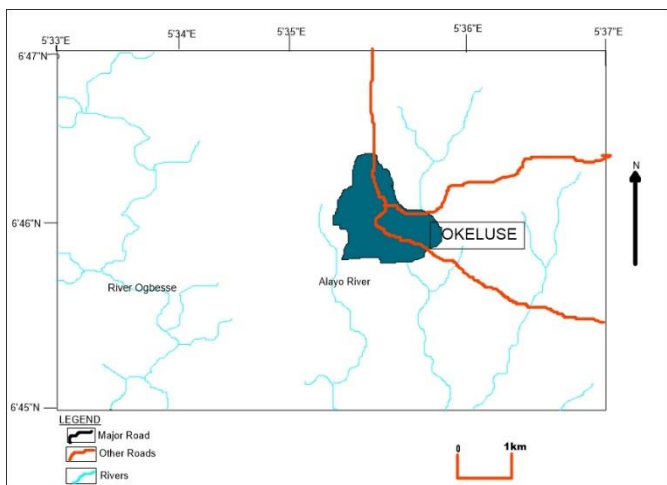


Figure 2: Map showing dendritic pattern of drainage in the study area

2. LOCAL GEOLOGY

The study area is underlain by Cretaceous sediments of Araromi formation belonging to Abeokuta group of Dahomey basin. Clayey and Lateritic topsoil generally covers the area. Due to undulating topography and its characteristic drainage pattern the exposed section was found along a stream channel in Okeluse. The lithologic units observed from the exposed section consists of dark grey thin fissile and carbonaceous shale (oldest) overlay by ferruginized sandstone having minor and marl occurrence follow by shallow operation of carbonaceous shale and clayey siltstone with the occurrence of mica flakes. Highly ferruginized sandstone showing ripples which are deposited on the alternating carbonaceous shale and clayey siltstone which overlapped the alternation of clay and white silt, having thick overburden that is densely vegetated at the top. Therefore, these lithologies can be grouped into benches which include topsoil (lateritic and clay), saturated clay followed by the limestone horizon (brownish fossiliferous limestone) and the fissile grey shale. The exposed lithologies at the site are made of four benches which are: the topsoil (lateritic and clay), the second bench which is saturated clay followed by the limestone horizon and the last bench is the fissile grey shale.

| Thickness (m) | Lithology | Description | Inferred Paleoenvironment |
|---------------|------------|--|---------------------------------------|
| | | Densely vegetated | |
| 5.36m | | Overburden | |
| 3.16m | | Alternation of Clay and white silt | Mud Flat |
| 1.96m | | Highly ferruginized, medium grained sandstone. Climbing ripples were observed towards the top | Tidal Flat |
| 0.9m | * C * B | Alternation of carbonaceous shale and clayey siltstone. Mica flakes were observed | Anoxic subtidal, reducing environment |
| 0.3m | | Parallel - bedded, medium grained, well sorted, consolidated, moderately hard, ferruginized sandstone with minor marl occurrence | Sand Flat |
| 0m | * A | Dark to grey, thin, fissile and carbonaceous shale | Anoxic and reducing environment |

Co - ordinates: Latitude N06°48' and Longitude E005°37'

* Sample Location

NOT DRAWN TO SCALE

Figure 3: Lithologic section of the Okeluse Limestone, S.W Nigeria

3. MATERIALS AND METHODS

Detailed geological mapping of Omi Alayo and Okeluse Forest Reserve was carried out by traversing and studying the exposed outcrops along the traverse. The outcrops were properly located on the field map and other observations were recorded in the field notebook. Ten (10) representative samples were collected using sledge hammer and were properly labeled using marker pens. These samples were taken from the exposed limestone deposits at varying intervals. Therefore, the sample numbers and coordinates of the locations were carefully noted in order to prevent mixing up of samples collected. Thereafter, all the samples were subjected to geochemical studies using X-ray Fluorescence Spectrophotometer (XRF) at the soil and water laboratory of the Federal Ministry of Water Resources, Ibadan, Nigeria. The main purpose of the analysis was to determine the major oxides (wt.%) and trace elements (ppm) in these rock samples. The laboratory methods to be used required that they must be sufficiently sensitive to detecting elements and be accurate for them to be useful in interpretation. First, the ten samples were crushed into smaller fragments and this was followed by grinding. The grinding was done in a ceramic mortar pestle by hand in order to ensure that they are

homogeneous. They were grinded into powdery form and packed into designated sample bags. The digestion and filtration of samples were done by total digestion method. The concentrations of Ni, Zn, Mn, Ba and Sr were also analyzed for on pressed pellets by X-Ray Fluorescence Spectrometry at same laboratory (operating conditions; Rh radiation, 70Kv, 40mA).

4. RESULTS AND DISCUSSION

The sampling points and their GPS coordinates are given in Table 1. Figure 4 shows the field photographs of limestone as exposed at Omi Alayo and Okeluse Forest Reserve (OFR). The results of geochemical analyses on the ten (10) representative samples from Omi Alayo (OA) and Okeluse Forest Reserve (OFR) are given in Tables 2 and 3. The geochemical analysis showed that CaO (lime) constitutes the bulk composition (Tables 2 and 3). The values ranged from 46.63 wt% to 49.73% wt % with mean value of 47.76 wt% for Omi Alayo limestone, and 47.76 wt% to 49.82 wt% with mean value of 48.89 wt% for the Okeluse limestone. The magnesium oxide (MgO) ranged from 0.70 wt% to 1.17 % with mean value of 0.83 wt% for Omi Alayo limestone, while it ranged from 0.56 wt% to 0.75 wt% with mean of 0.69 wt% for the Okeluse limestone.

For the Al₂O₃, it varied from 1.06% to 2.60% wt% with mean value of 1.84 wt% for Omi Alayo and 1.16 wt% to 2.74 wt% with mean of 2.00wt% for Okeluse deposits (Tables 2 and 3). In case of the SiO₂ content, it ranged from 6.10 wt% to 8.75 wt% with the mean value of 7.53 wt% for Omi Alayo limestone and 6.16 wt% to 8.46 wt% with mean of 7.03 wt% for Okeluse samples. Other oxides; Na₂O, P₂O₅, K₂O and TiO₂ are low in concentration with means values of 0.43, 0.00, 0.56, and 0.15 wt% for Omi Alayo and 0.29, 0.00, 0.46 and 0.15 wt% for Okeluse limestone (Tables 2 and 3). Generally, the loss on the ignition (LOI) of the limestone deposits have values less than 40% with values of 38.70 and 38.68 wt% for all locations (Tables 2 and 3).

| Table 1: Showing the sampling points and the GPS coordinates of Okeluse limestone | | |
|---|--------------|---------------|
| Sampled points | Latitude (N) | Longitude (E) |
| L1 - Omi-Alayo Limestone (OA) | 06° 47. 07' | 05° 35. 02' |
| L2 - Okeluse Forest Reserve (OFR) Limestone | 06° 47.24' | 05° 36. 50' |



Figure 4: Field photographs of limestone deposits at Omi Alayo and Okeluse Forest Reserve, SW Nigeria

| Table 2: Major oxides (wt %) of Omi Alayo limestone | | | | | | | | |
|---|-------|-------|-------|-------|-------|-----------|------------|-------|
| Sample No. Oxide(wt%) | OA1 | OA2 | OA3 | OA4 | OA5 | Range Low | Range High | Mean |
| SiO ₂ | 8.55 | 6.10 | 6.50 | 7.75 | 8.75 | 6.10 | 8.75 | 7.53 |
| TiO ₂ | - | 0.15 | - | - | - | - | - | 0.15 |
| Al ₂ O ₃ | 2.00 | 2.60 | 1.43 | 1.06 | 2.10 | 1.06 | 2.60 | 1.84 |
| Fe ₂ O ₃ | 2.32 | 1.10 | 1.65 | 1.06 | 2.30 | 1.60 | 2.32 | 1.69 |
| MgO | 1.14 | 0.70 | 0.74 | 0.71 | 0.90 | 0.70 | 1.14 | 0.83 |
| MnO | - | - | - | - | - | - | - | - |
| K ₂ O | 0.91 | 0.63 | 0.17 | 0.12 | 0.95 | 0.12 | 0.95 | 0.56 |
| Na ₂ O | 0.98 | 0.06 | 0.18 | 0.09 | 0.85 | 0.06 | 0.98 | 0.43 |
| CaO | 46.63 | 47.56 | 49.73 | 48.09 | 48.65 | 46.63 | 49.73 | 48.10 |
| P ₂ O ₅ | - | - | - | - | - | - | - | - |
| LOI | 37.47 | 41.07 | 39.60 | 40.15 | 35.24 | 35.24 | 41.07 | 38.70 |
| Total | 100.0 | 99.97 | 100.0 | 98.94 | 99.74 | 91.51 | 107.52 | 99.83 |

OA - Omi - Alayo

Table 3: Major oxides (wt %) of Okeluse Forest Reserve limestone (OFR)

| Sample No. Oxide (wt.%) | OFR 6 | OFR 7 | OFR 8 | OFR 9 | OFR 10 | Range Low | Range High | Mean |
|--------------------------------|--------|-------|--------|-------|--------|-----------|------------|-------|
| SiO ₂ | 6.16 | 7.80 | 6.55 | 8.46 | 6.18 | 6.16 | 8.46 | 7.03 |
| TiO ₂ | 0.17 | 0.14 | - | - | 0.15 | 0.14 | 0.17 | 0.15 |
| Al ₂ O ₃ | 2.58 | 1.16 | 1.48 | 2.04 | 2.74 | 1.16 | 2.74 | 2.00 |
| Fe ₂ O ₃ | 1.52 | 0.56 | 1.58 | 2.42 | 1.12 | 0.56 | 2.42 | 1.44 |
| MgO | 0.72 | 0.56 | 0.75 | 0.71 | 0.72 | 0.56 | 0.75 | 0.69 |
| MnO | - | - | - | - | - | - | - | - |
| K ₂ O | 0.52 | 0.16 | 0.16 | 0.82 | 0.65 | 0.16 | 0.82 | 0.46 |
| Na ₂ O | 0.05 | 0.09 | 0.28 | 0.99 | 0.08 | 0.05 | 0.99 | 0.29 |
| CaO | 49.56 | 48.42 | 49.82 | 48.76 | 47.88 | 47.76 | 49.82 | 48.89 |
| P ₂ O ₅ | - | - | - | - | - | - | - | - |
| LOI | 38.72 | 39.49 | 39.38 | 35.34 | 40.48 | 35.34 | 40.48 | 38.68 |
| Total | 100.00 | 98.38 | 100.00 | 99.54 | 100.00 | 91.89 | 106.65 | 99.63 |

OFR- Okeluse Forest Reserve

From the result of elemental oxides, CaO is predominant compared to other oxides analysed. There is little difference in the CaO contents of Omi Alayo and Okeluse limestone deposits. Also, high value of CaO corresponds to low values of SiO₂ and MgO. This is an indication of purity of limestone in these areas. The CaO of Omi-Alayo and Okeluse compared favourably with those obtained for Nkalagu (49.74), Gboko (48.88) but fell short of Sagamu (89.20) and Ewekoro (80.3) (Ikhane et al.,2009; Bejide, 2000; Akinmosun et al.,2005). The silica content of studied limestone are relatively similar to those Sagamu and Gboko, but higher than those of pure limestone of Pettijohn (Table 4) (Pettijohn, 1975). This study considered the limestone deposits of Omi Alayo and Okeluse as low silica limestone and of high purity. Generally, the limestone deposits in the

two locations are low in Alumina and Magnesium contents. Mean values of 2.00 and 1.84 for Alumina are obtained for the deposits at Omi Alayo and Okeluse respectively. Also, MgO mean values ranged from 0.56 to 0.60 (Tables 2 and 3). Low amount of alumina is an indication of deposition in low energy environment. A group researchers had reported presence of brachiopods, gastropods, bivalves and ostracods in some limestones of Arimogija – Okeluse limestone which obviously indicate an open shelf environment (Ehinola et al., 2012). Pettijohn opined that magnesium content is a function of temperature and often low in shells living in shallow waters (Pettijohn, 1975). It can be inferred that the limestone of Omi Alayo and Okeluse are of shallow marine environment. In an attempt to classify the

Table 4: Result of major oxides from other limestone deposits in Nigeria

| | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | MgO | CaO | Na ₂ O | K ₂ O | TiO ₂ | P ₂ O ₅ | LOI |
|----|------------------|--------------------------------|--------------------------------|------|------|-------------------|------------------|------------------|-------------------------------|------|
| 1. | 1.88 | 0.83 | 0.26 | 2.75 | 50.8 | 0.06 | 0.01 | 0.01 | 0.01 | - |
| 2 | 7.28 | 1.92 | 1.56 | 0.76 | 48.5 | 0.36 | 0.51 | 0.15 | - | 99.7 |
| 3 | 5.90 | 1.30 | 0.77 | 0.99 | 49.7 | - | - | - | - | - |
| 4 | 9.78 | 1.48 | 2.20 | 1.99 | 48.8 | 0.05 | 0.01 | - | - | 36.5 |
| 5 | 5.70 | - | - | - | 89.2 | - | - | - | - | - |

1. Pure Limestone (Pettijohn, 1975)
2. This Study (average value for Omi-Alayo and Okeluse)
3. Nkalagu (Ikhane et al., 2009)
4. Gboko Limestone (Bejide, 2000)
5. Sagamu Limestone (Akinmosun et al.,2005)

limestone of these areas, after Todd, the ratios of Ca/Mg and Mg/Ca was obtained for them (Todd, 1966). Todd classified limestone samples with Ca/Mg ratio with range 1.14% - 12.30% as dolomitic limestone, samples with Ca/Mg of 12.30% - 39.00% as "Magnesian limestone" and samples with 39% - 100% as 'Pure limestone' (Table 5) (Todd, 1966). The reciprocal Mg/Ca ratio varies from 25.87 to 37.63, while Mg/Ca ranges from 0.03 - 0.04. In this study, the limestone samples from Omi-Alayo and Okeluse Forest Reserve can be classified as Pure Limestone type (Table 6). Marshner had reiterated that Ca/Mg ratio corresponds to stability condition during formation of carbonate rocks (Marshner, 1968). He went further to relate the degree of salinity with the Ca/Mg ratio and concluded that the degree of salinity increases with decrease in Ca/Mg ratio. Hence, in this study less evaporation of sea water corresponds to low salinity that

prevailed when the limestone were deposited. For the Loss of ignition (LOI), average value of 38.69 was obtained for these 10 locations. LOI indicate the content of volatiles available in any limestone deposit. High amount of LOI suggest high volatile content and invariably an indication of carbonate content which is also the release of carbon dioxide after heating up to 1000°C (Olatunji,1989). The Trace element concentrations of the limestone studied are reported in ppm (Table 7) has Barium (Ba) ranged from 15 to 25ppm with an average of 19.5ppm. Strontium (Sr) ranged from 400ppm to 510ppm with an average of 458.2ppm, while Zircon (Zr) ranged from 8ppm to 36ppm with an average of 21.8ppm. Trace element compositions are useful in

Table 5: Classification of Limestone (after Todd, 1966)

| Descriptive term | Standard Ratio Ca/Mg | Reciprocal Ratio Mg/Ca |
|---------------------|----------------------|------------------------|
| Dolomitic Limestone | 12.30 - 1.41 | 0.08 - 0.18 |
| Magnesian Limestone | 39.00 - 12.30 | 0.03 - 0.08 |
| Pure Limestone | 100.00 - 39.00 | 0.00 - 0.18 |

Table 6: Chemical classification of Omi-Alayo and Okeluse Limestone

| Sample No. | MgO (w%) | CaO (wt%) | Ca/Mg | Mg/Ca | Remarks |
|------------|----------|-----------|-------|-------|----------------|
| OA 1 | 1.14 | 46.63 | 40.90 | 0.024 | Pure Limestone |
| OA 2 | 0.70 | 47.56 | 67.94 | 0.014 | Pure Limestone |
| OA 3 | 0.74 | 49.73 | 67.20 | 0.014 | Pure Limestone |
| OA 4 | 0.71 | 48.09 | 67.73 | 0.014 | Pure Limestone |
| OA 5 | 0.90 | 48.65 | 54.05 | 0.018 | Pure Limestone |
| OFR 6 | 0.72 | 49.65 | 68.95 | 0.014 | Pure Limestone |
| OFR 7 | 0.56 | 48.42 | 86.46 | 0.011 | Pure Limestone |
| OFR 8 | 0.75 | 49.82 | 66.42 | 0.015 | Pure Limestone |
| OFR 9 | 0.71 | 48.76 | 68.67 | 0.014 | Pure Limestone |
| OFR 10 | 0.72 | 47.88 | 66.50 | 0.015 | Pure Limestone |

Table 7: Trace element composition (ppm) of Omi-Alayo- Okeluse Limestone

| Sample No. | Ba | Sr | Zr |
|------------|------|-------|------|
| OA1 | 25 | 501 | 32 |
| OA2 | 22 | 510 | 33 |
| OA3 | 20 | 502 | 35 |
| OA4 | 24 | 500 | 36 |
| OA5 | 25 | 508 | 35 |
| OFR6 | 16 | 400 | 10 |
| OFR7 | 18 | 413 | 8 |
| OFR8 | 15 | 408 | 10 |
| OFR9 | 15 | 410 | 9 |
| OFR10 | 15 | 430 | 20 |
| Mean | 19.5 | 458.2 | 21.8 |

Determination of depositional environment. A veritable tool in discriminating shallow water from deep water limestones. Carbonates are noted for enrichment in strontium (Sr^{2+}) due to the fact that they readily substitute for calcium (Ca^{2+}). Flugel and Wedepohl had reported low Sr values (100-400ppm) for shallow marine and high strontium values for (Oberalm limestone) for deep sea water carbonates (Flugel and Wedepohl, 1967). Ofulume, also reported 100 -400ppm for strontium and high strontium values for shallow water and deep-water carbonates (Ofulume, 2012). The strontium (Sr) values in Omi-Alayo-Okeluse limestone indicate that they were deposited under higher saline/ shallow water environmental conditions.

5. CONCLUSION

Limestone is an important component in the manufacture of cement worldwide. They are quarried in different parts of Nigeria. However their suitability for cement production depends on its chemical composition. This helps in determining their purity. In this study, ten (10) representative samples were collected in five locations around Om-Alayo and Okeluse forest reserve located in the Eastern Dahomey Basin, southwestern Nigeria. The samples were subjected to geochemical analyses and the results showed that all samples are enriched in CaO relative to other major oxides. Mean values of 48.10 wt% and 48.89 wt% recorded for Omi-Alayo and Okeluse limestones respectively. Inverse relationship between CaO and SiO_2 contents suggest its suitability for cement production. The Todd classification scheme for limestone showed that the limestone in both locations classified as Pure Limestone. Ca/Mg ratio ranged from 40.90 to 86.46 and reciprocal ratio Mg/Ca range of 0.011 - 0.024. Trace amount of alumina and magnesium predicts shallow waters for the limestone formation in this study. Trace element composition, also showed that the formation of Omi-Alayo-Okeluse limestone to be under shallow water conditions. In conclusion, the limestone in both locations are pure limestone type and suitable for cement production.

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