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Geochemical properties of limestone and its raw material potential for cement manufacturing in the Ras Bayad area of northeastern Libya

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الخصائص الجيوكيميائية للحجر الجيري وإمكانات المواد الخام لصناعة الإسمنت في منطقة رأس بياض بشمال شرق ليبيا

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Abstract

The purpose of this study was to examine the geochemical characterizations of the limestone and clay deposits in the Ras Bayad area in order to classify and purify them as well as evaluate their potential as raw materials for the production of cement. Using the X-ray fluorescence analysis (XRF) technique, fifteen representative rock samples were collected from three chosen locations for both carbonate and clay rocks. The results of chemical analyses have been used to create ratio factors analysis models such as silica modulus (SM), alumina modulus (AM), hydraulic modulus (HM), lime saturation factor (LSF), and liquid phase (L. phase). Additionally, Bogue's formulas have been used to calculate quality control coefficients like C2S, C3S, C3A, and C4AF. The obtained results showed that the raw materials under consideration show that the geochemical classification according to the Al2O3–CaO–(MgO+FeOt) ternary diagram and Ca/Mg ratio as well as CaO%, CaCO3% contents are pure limestones and can be used for cement manufacture due to the high lime (CaO%) content and low impurity content. Lime saturation factor (LSF), silica modulus (SM), and alumina modulus (AM) ratio factor analysis demonstrated that these rocks met these factors' requirements. The calculated different moduli and quality control coefficients, such as C2S, C3S, C3A, and C4AF, showed that there are some differences between different rock types and locations. This is typically due to the chemical composition of each type, which is mainly dependent on the nature of depositional under specific geological conditions. The geochemical suitability of limestone demonstrated that these raw materials meet standard requirements for cement manufacturing. The raw material blending ratios for the limestone and clay rock samples at locations (1), (2), and (3) are 1:2.38, 1:2.47, and 1:2.44, respectively, with minimal variation.

Keywords: Geochemistry, limestone, clay, raw material, assessment, cement, quality models, bending ratio.

الملخص

هدفت هذه الدراسة إلى فحص الخصائص الجيوكيميائية لرواسب الحجر الجيري والطين في منطقة رأس بياد، وذلك لتصنيفها وتنقيتها، بالإضافة إلى تقييم إمكانية استخدامها كمواد خام لإنتاج الإسمنت. وباستخدام تقنية تحليل الأشعة السينية الفلورية (XRF)، جُمعت خمس عشرة عينة صخرية ممثلة من ثلاثة مواقع مختارة، شملت كلاً من الصخور الكربوناتية والطينية. استُخدمت نتائج التحليلات الكيميائية لإنشاء نماذج تحليل عوامل النسب، مثل معامل السيليكا (SM)، ومعامل الألومينا (AM)، والمعامل الهيدروليكي (HM)، ومعامل تشبع الجير (LSF)، والطور السائل (الطور السائل). بالإضافة إلى ذلك، استُخدمت معادلات بوغ لحساب معاملات مراقبة الجودة، مثل C2S وC3S وC3A وC4AF. أظهرت النتائج أن المواد الخام قيد الدراسة تُصنف جيولوجيًا وفقًا للمخطط الثلاثي (Al2O3–CaO–(MgO+FeOt ونسبة Ca/Mg، فضلًا عن محتوى CaO% وCaCO3%، على أنها أحجار جيرية نقية، ويمكن استخدامها في صناعة الأسمنت نظرًا لارتفاع محتوى الجير (CaO%) وانخفاض محتوى الشوائب. وقد أثبت تحليل عوامل نسب معامل تشبع الجير (LSF) ومعامل السيليكا (SM) ومعامل الألومينا (AM) أن هذه الصخور تستوفي متطلبات هذه العوامل. أظهرت معاملات التحكم في الجودة المختلفة المحسوبة، مثل C2S و C3S و C3A و C4AF، وجود بعض الاختلافات بين أنواع الصخور المختلفة والمواقع. يعود هذا عادةً إلى التركيب الكيميائي لكل نوع، والذي يعتمد بشكل أساسي على طبيعة الترسيب في ظل ظروف جيولوجية محددة. وقد أظهرت الملاءمة الجيوكيميائية للحجر الجيري أن هذه المواد الخام تفي بالمتطلبات القياسية لصناعة الإسمنت. وكانت نسب خلط المواد الخام لعينات الحجر الجيري والصخور الطينية في المواقع (1) و(2) و(3) هي 1:2.38 و1:2.47 و1:2.44 على التوالي، مع اختلاف طفيف.

الكلمات المفتاحية: الجيوكيمياء، الحجر الجيري، الطين، المواد الخام، التقييم، الإسمنت، نماذج الجودة، نسبة الانحناء.

1. INTRODUCTION

It is frequently claimed that limestone is the most adaptable mineral in the world and is an essential raw material. Although it has several uses, its main one is in the building sector, where it is the main supplier of crushed rock aggregate in many nations. It is also a source of building stones and a necessary raw material for the production of cement. As seen in Figure 1, the term "industrial limestone" refers to limestone used for non-constructional uses where its chemical characteristics or level of whiteness are crucial.

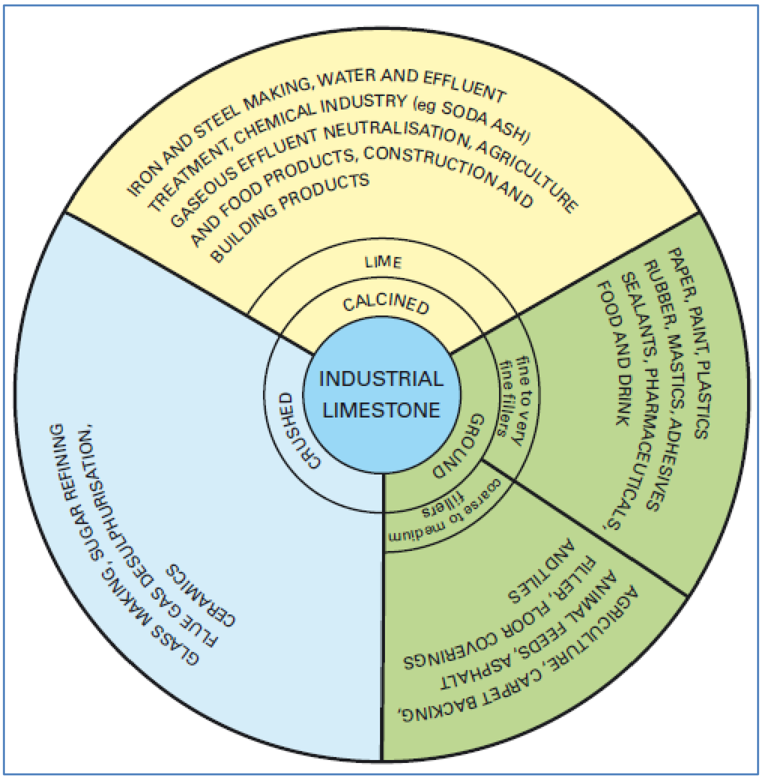


Figure 1: Industrial uses of limestone.

The process of determining a substance's chemical composition, concentration, and other chemical characteristics in order to diagnose it is known as geochemical analysis.
The content of an element or combination of elements is determined through chemical analysis. Chemical examinations of the material are crucial to the geological study of mineral resources [1]. Rock and other naturally occurring materials are the subject of the geochemical investigation. One of the significant industrial rocks that is currently taking the lead in the production of cement worldwide is limestone, which is said to be the primary raw material needed for the cement industry [2,3].
based on a study [4] that looked at how limestone composition affected Portland-limestone cement's (PLC) performance. Two distinct clinkers were interground with three limestones, one of which contained calcite with a greater calcium carbonate concentration and the others mostly dolomite. They found that the limestone offered comparable or better strength and rate of strength development for all materials and grinding periods tested at up to 15% limestone addition.
As the population has grown, so has the need for limestone. It can form from the shells of dead sea creatures (bioclastic limestone), be secreted by marine organisms like algae and coral (biochemical limestone), or precipitate from water (non-clastic, chemical, or inorganic limestone). Certain limestones, which frequently resemble sandstone or mudstone, are created by cementing sand and/or mud using calcite (clastic limestone). Limestone will fizz in diluted hydrochloric acid because calcite is its primary mineral component. In order to assess the purity and suitability of limestone for industrial uses, this study describes the chemical composition and classifications of limestone found in the studied area [5].

2. LOCATION OF STUDY

This study has been carried out on Ras Bayad area which located at the coast of Mediterranean Sea in the northeastern of Libyan as shown in Figure 1.

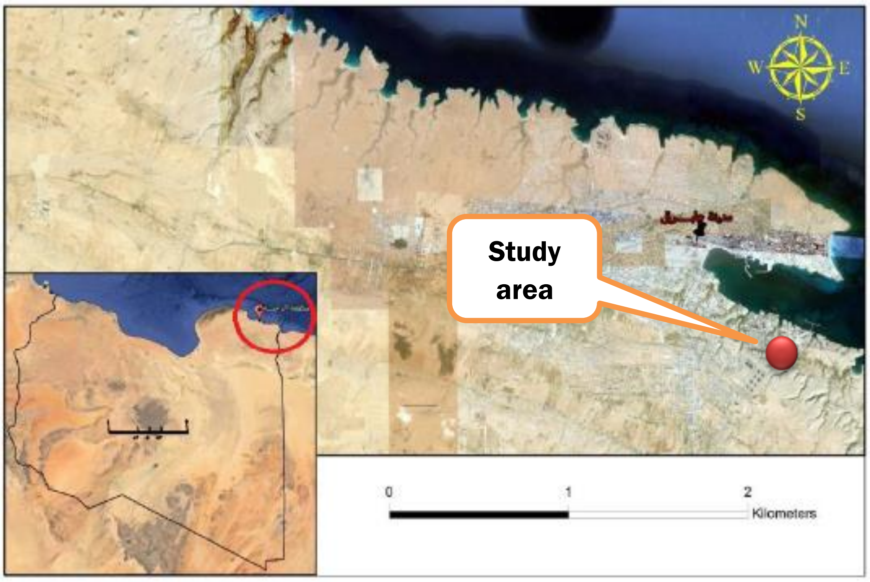


Figure 2: Satellite map depicts the location of Ras Bayad area

3. STUDY OBJECTIVES

The main aims of this study is to focus on the assessment of the raw materials required for cement industry in the investigated locations throughout:

1. Study the chemical composition of raw materials particularly limestone and clay.
2. Determination the potentiality for cement manufacture.
3. Calculation the appropriate blending ratios of mix.
4. Highlight on the raw materials as natural resources in Libya.
5. Correlation between these raw materials and the standard ones to distinguish their quality for cement clinker.

4. STUDY SIGNIFICANCE

The importance of this study can be summarized as:

1. Libya has a great quantities of raw materials estimated as billions of tons.
2. The raw materials characterized by the lower contents of impurities.
3. The most locations of these materials located on or nearby the highways.
4. The easy of transportation by trucks or belt conveyor.
5. To achieve the sustainable development and support the national income.

5. METHODOLOGY

To assessment the quality of the raw materials in the studied areas for cement manufacture, samples were collected from three locations namely Location (1), Location (2) and Location (3). Fifteen limestone rock samples have been collected, five samples for each locations. These samples are denoted by LS1,1 till LS1,5; LS2,1 till LS2,5 and LS3,1 till LS3,5 for the three locations respectively.

These samples are subjected to chemical analyses in Libyan National Cement Company at Al Fataih area to determine the different constituents using XRF instrument.

To assessment the potential of utilize these rocks as raw materials for cement manufacture, the following models were applied:

1.Hydraulic Modulus (HM)
2.Lime Saturation Factor (LSF)
3.Silica Modulus (SM)
4.Alumina Modulus (AM)

Also, the quality control coefficients of raw materials have been estimated based on the calculations of apparent values of clinker constituents such as C2S, C3S, C3A and C4AF using Bogue's formulae, that rely on the typical chemical composition of limestone rocks suggested by different authors.

6. GEOLOGICAL SETTING

One of the most prevalent sedimentary rocks in the study region is limestone, which covers a sizable portion of Libya. It is a thick succession of mostly calcareous sediments with notable sequences of different kinds of limestone present locally. This region's limestone resources are distinguished by its large quantities, high quality, and placement along the Mediterranean Sea coast.

6.1. Lithology and Stratigraphy

Carbonates, calcareous fossiliferous rocks, and clay deposits are among the few rock types that generally exhibit the defining lithology of the examined location. The thickness and extent of the stratigraphic succession vary from place to place as a result of tectonic movements and variations in depositional variables.

Three primary Early Tertiary stratigraphic formations are identified in the thick sedimentary section exposed in the Ras Bayad region, particularly along the cliffs facing the Mediterranean Sea as seen in Figures 2 to 3. In the field, the following units were identified as follows:

Figure 3: Exposure limestone in Location (1) at coastal area

Figure 4: Limestone deposits in Location (2 left) and Location (3 right).

Figure 5: Location of clay deposits.

Quaternary

•Represented by alluvium deposits, Beach and coastal sand dune and Eolian deposits.

Tertiary

•Lower-Middle Miocene: Al Jaghbub Formation

•Upper Oligocene-Lower Miocene: Al Faidiyah Formation

•Upper Eocene-Lower Oligocene: Al Khowaymat Formation

Al Faidiyah Formation: white limestone, faint brown to dark yellow fossiliferous limestones and sometimes intercalated with clay deposits. The beds are nearly horizontal, thin to thin-bedded and highly fossiliferous. It is including the following assemblage of macrofauna: Eoconoids, Econoidea, Mollusca Pelecypoda (Figure 6).

Figure 6: Fossiliferous limestone of Al Faidiyah Formation

Al Khowaymat Formation: dolomitic limestone, yellowish white, hard compact, fossiliferous including ; Globgerina spp., Globorotalia spp. and Nummulites.
Al Jaghbub Formation: marly limestone, dark yellow and moderately hard. It is including the following assemblage of macrofauna: Ostreaverleti, Ostreadigitalina, Cardiumerinaceum and Pectencristato.

6.2. Limestone and Clay Raw Materials

Raw materials samples of limestone and clay rocks were collected from locations under consideration of limestone deposits and chemically analysed for major oxides as additive raw materials for cement industry.

The investigation study has been carried out through a selected areas due to several considerations that can be categorized as following:

1. The quality of limestone rocks.
2. The region's location on the Mediterranean Sea coast makes it easier to move goods and raw materials.
3. The area's paved roadways.
4. The enormous limestone rock reserves.
5. Infrastructure accessibility.

7. RESULTS AND DISCUSSION

7.1. Geochemical Analysis

Tables 1, 2 and 3 provide the chemical analyses of limestone rock samples from the three locations of the study, in addition to their ranges and averages. On the other hand, the averages chemical analyses of clay rock samples are presented in Table 4.

Table 1 Chemical composition of limestone rocks at location (1)

Components (%)	Location (1)						
	SL1,1	SL1,2	SL1,3	SL1,4	SL1,5	Range	Average
SiO_2	0.00	0.03	0.00	0.02	0.04	0.00-0.04	0.018
Al_2O_3	0.13	0.12	0.15	0.18	0.10	0.10-0.18	0.136
Fe_2O_3	0.00	0.00	0.02	0.01	0.00	0.00-0.02	0.006
CaO	55.92	54.80	54.51	55.01	55.72	54.51-55.92	55.19
MgO	0.52	0.45	0.44	0.51	0.56	0.44-0.56	0.50
Cl	0.08	0.07	0.10	0.02	0.09	0.07-0.10	0.07
SO_3	0.13	0.10	0.12	0.14	0.19	0.10-0.19	0.14
Na_2O	0.00	0.001	0.00	0.002	0.00	0.00-0.02	0.001
K_2O	0.00	0.002	0.001	0.00	0.00	0.00-0.02	0.001
TiO_2	0.00	0.001	0.002	0.00	0.001	0.00-0.02	0.001
MnO	0.01	0.00	0.01	0.02	0.03	0.00-0.03	0.01
P_2O_5	0.19	0.12	0.10	0.20	0.24	0.10-0.24	0.17
LOI*	42.96	44.16	44.18	43.70	42.95	42.95-44.18	43.59
$\sum$	99.94	99.85	99.63	99.82	99.92	99.63-99.94	99.83

LOI* = loss on ignition

Table 2 Chemical composition of limestone rocks at location (2)

Components (%)	**Location (2)**						
	SL2,1	**SL2,2**	**SL2,3**	**SL2,4**	**SL2,5**	**Range**	**Average**
SiO_2	0.02	0.03	0.00	0.05	0.01	0.00-0.05	0.02
Al_2O_3	0.14	0.19	0.18	0.14	0.12	0.12-0.19	0.15
Fe_2O_3	0.01	0.00	0.02	0.03	0.01	0.00-0.03	0.01
CaO	54.95	55.80	53.51	54.01	53.72	53.51-55.80	54.40
MgO	0.50	0.40	0.74	0.58	1.65	0.40-1.65	0.77
Cl	0.00	0.06	0.12	0.02	0.08	0.00-0.12	0.06
SO_3	0.12	0.11	0.13	0.10	0.14	0.10-0.14	0.12
Na_2O	0.00	0.001	0.001	0.003	0.001	0.00-0.003	0.001
K_2O	0.00	0.002	0.002	0.00	0.001	0.00-0.002	0.001
TiO_2	0.01	0.002	0.001	0.00	0.001	0.00-0.001	0.003
MnO	0.01	0.00	0.02	0.01	0.01	0.00-0.02	0.01
P_2O_5	0.20	0.13	0.10	0.18	0.20	0.10-0.20	0.16
LOI*	43.87	43.16	44.18	44.70	43.95	43.16-44.70	43.97
∑	99.83	99.89	99.01	99.82	99.89	99.01-99.98	99.69

LOI* = loss on ignition

Table 3 Chemical composition of limestone rocks at location (3)

Components (%)	Location (3)						
	SL3,1	SL3,2	SL3,3	SL3,4	SL3,5	Range	Average
SiO_2	0.01	0.04	0.00	0.05	0.09	0.00-0.09	0.04
Al_2O_3	0.15	0.19	0.20	0.12	0.18	0.12-0.20	0.17
Fe_2O_3	0.00	0.001	0.02	0.01	0.01	0.00-0.02	0.008
CaO	55.19	54.95	53.52	54.34	53.90	53.52-55.19	54.38
MgO	0.54	0.90	1.74	0.58	1.60	0.54-1.74	1.07
Cl	0.06	0.02	0.12	0.04	0.10	0.02-0.12	0.07
SO_3	0.14	0.12	0.17	0.19	0.11	0.11-0.19	0.15
Na_2O	0.00	0.00	0.001	0.002	0.001	0.00-0.002	0.001
K_2O	0.00	0.003	0.002	0.00	0.001	0.00-0.003	0.002
TiO_2	0.00	0.004	0.001	0.003	0.001	0.00-0.004	0.002
MnO	0.02	0.00	0.03	0.01	0.02	0.00-0.03	0.02
P_2O_5	0.21	0.09	0.10	0.17	0.21	0.09-0.21	0.16
LOI*	43.62	43.16	44.02	44.32	43.45	43.16-44.32	43.71
∑	99.94	99.48	99.92	99.85	99.67	99.48-99.94	99.77

LOI* = loss on ignition

Table 4 Chemical composition of clay rocks

Components (%)	Location (1)	Location (2)	Location (3)	Average
SiO_2	36.55	37.10	37.06	36.90
Al_2O_3	7.9	8.09	7.98	7.99
Fe_2O_3	4.1	4.05	4.15	4.10
CaO	20.1	21.08	21.40	20.86
MgO	3.34	3.30	3.25	3.30
Cl	0.81	0.80	0.75	0.77
SO_3	1.25	1.30	1.32	1.29
Na_2O	0.27	0.30	0.24	0.27
K_2O	0	0.00	0.01	0.003
TiO_2	0.01	0.02	0.00	0.010
MnO	0.02	0.01	0.02	0.017
P_2O_5	0.15	0.14	0.17	0.15
LOI*	25.49	23.72	23.50	24.24
$\sum$	99.99	99.91	99.85	99.92
$CaCO_3$	63.40	62.50	61.45	62.45
$MgCO_3$	40.30	42.44	41.95	41.56

LOI* = loss on ignition

7.2. Geochemical Constituents

The principal oxide elements' geochemical distribution may directly reveal details about the depositional environment. Tables 1, 2, and 3 present the main oxide data for the three sites for the chosen limestone samples that were examined using the X-ray fluorescence technique at Libyan National Cement Company. There is no compositional heterogeneity in any of the main oxide constituents. In contrast, the research sites (1), (2), and (3) had CaO averages of 55.19%, 54.40%, and 54.38%, respectively. Geochemical examination of the limestone samples revealed that the site (1) had the most CaO (55.19%) and the lowest silica (0.018%) because of the presence of calcite. Increased MgO concentration in limestones increases the dolomite component and, under favourable circumstances, may intensify alkali carbonate reactions. "Low Mg-calcite" (LMC) refers to limestones with less than 4% MgCO3, whereas "high Mg-calcite" (HMC) refers to limestones with more than 4% MgCO3. Less than 5% MgO by volume is needed for applications such as dimension stone, cement, roadstone, and concrete. Because of the lower MgO content averages (0.50%, 0.77%, and 1.07% for sites (1), (2), and (3), respectively), the limestones under evaluation are deemed low Mg-calcite and acceptable.

However, the low levels of Al_2O_3, K_2O, Na_2O, and SO_3 related to the decreasing spread of clays. MgO contents are extremely low content indicates a weak dolomitization process of the limestone.

Plotting limestone samples on the Al_2O_3–CaO–(MgO + FeO_t) ternary diagram shows that all samples are rather pure limestones as shown in Figure 7.

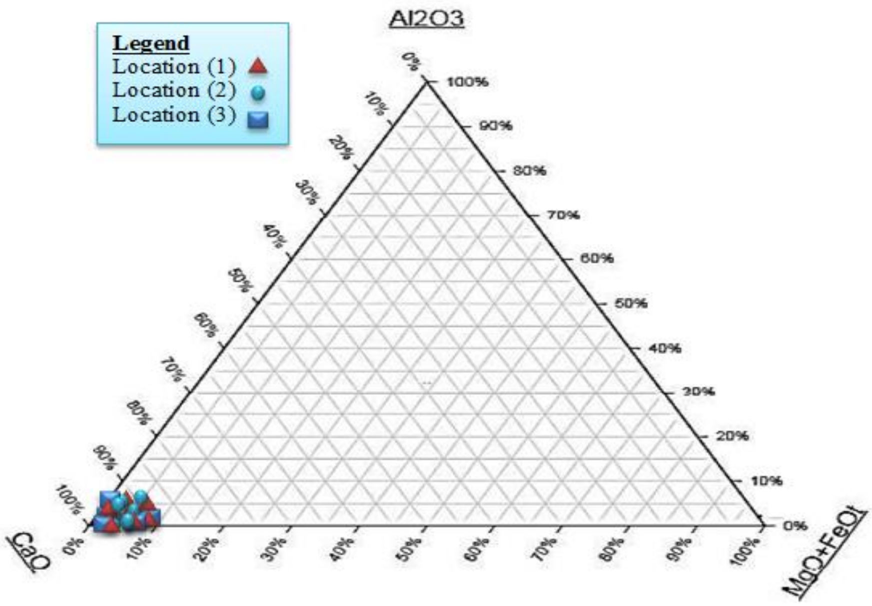


Figure 7: Representation of limestone on Al_2O_3–CaO–(MgO + FeO_t) ternary diagram

7.3. Geochemical Classification

For pure limestone, Todd's (1966) standard Ca/Mg and Mg/Ca ratios fall between 100-39.0 and 0-0.3, respectively; for magnesian limestone, they are between 38.0-12.3 and 0.03-0.08, respectively; and for dolomitic limestones, they are between 12.30-1.41 and 0.08-0.18. It is discovered that the distribution of Ca/Mg and its reciprocal Mg/Ca ratios in the limestone of the research areas varies from place to place (Table 6). These rocks can be categorised as pure limestone based on the ratios and metrics Todd [6] used. The stability condition during the carbonate rock's creation is correlated with the Ca/Mg ratio. Mashner [7] noted that as the Ca/Mg ratio decreases, the degree of salinity rises. Higher Ca/Mg ratios in the carbonates under study are indicative of low salinity and relatively less seawater evaporation in the overall limestone formation. All of the limestone samples, however, meet Todd's criteria [6].

Table 6 Chemical classification of limestone rocks using Todd's classification method

Location (1)						
Elements ratio	SL1,1	SL1,2	SL1,3	SL1,4	SL1,5	Range
Ca/Mg	107.54	121.78	123.89	107.86	99.50	99.50-123.89
Mg/Ca	0.009	0.008	0.008	0.009	0.010	0.008-0.10
Limestone classification	Pure limestone rocks					
Location (2)						
Elements ratio	SL2,1	SL2,2	SL2,3	SL2,4	SL2,5	Range
Ca/Mg	109.90	139.50	72.31	93.12	32.56	32.56-139.50
Mg/Ca	0.009	0.007	0.014	0.011	0.031	0.007-0.031
Limestone classification	Pure limestone rocks					
Location (3)						
Elements ratio	SL3,1	SL3,2	SL3,3	SL3,4	SL3,5	Range
Ca/Mg	102.20	61.06	30.76	93.69	33.69	30.76-102.20
Mg/Ca	0.010	0.016	0.033	0.011	0.030	0.010-0.033
Limestone classification	Pure limestone rocks					

The geochemical analysis's findings above indicate that limestone may be used in Portland cement. One of the main basic materials needed to make Portland cement is limestone [8]. Sulphur should be minimal, phosphorous pentaoxide should be less than 0.5%, and magnesia should be less than 3.0% in the chemical standard. As a result, these limestones fit the aforementioned specifications and don't require beneficiation to satisfy the needs of the cement industry, allowing for direct use in the production of cement.
However, according to the authors [9,12], limestones can be categorised as follows based on carbonate concentration: > 98.5% CaCO3 very high purity, 97.0 to 98.5% high purity, 93.5 to 97.0% medium purity, 85.0 to 93.5% low purity, and 50.0 to 85.0% impure (Table 7). Carbonate rock containing more than 98.5% calcium carbonate (typically as calcite) is referred to as very high purity limestone. In all significant limestone resource research conducted in the UK and other regions of the world, the British Geological Survey (BGS) has employed this categorisation [13].

Table 7 Limestone purity classification according to [9,12]

Purity classification	$CaCO_3$ wt.%	CaO wt.%	Possible industrial uses (grouped by minimum $CaCO_3$ specifications)
Very high purity	> 98.5	> 55.2	Steel, white glass (subjected to trace elements), rubber, plastics, paint
High purity	97.0-98.5	54.3-55.2	High purity 97.0–98.5 54.3–55.2 Iron, ceramic, Portland cement, whiting, chemical uses
Medium purity	93.5-97.0	52.4-54.3	Medium purity 93.5–97.0 52.4–54.3 Paper (subject to color), animal feedstuffs (subjected to level
Low purity	85.0–93.5	47.6–52.4	Asphalt
Impure	< 85.0	< 47.6	Mineral wool, natural cements (subjected to silica/clay mineral ratio)

Based on the obtained chemical analyses that presented in Tables 1,2 & 3, and according to CaO% in addition to the $CaCO_3$%, that are ranging from 98.12-98.82% (averaging 98.40%), 97.90-98.82% (averaging 98.41%) and 97.10-98.43% (averaging 98.03%) for Locations (1), (2) and (3) respectively, it could be classified as high purity limestones. Some limestone samples exhibit very high purity limestones (Table 9).

Table 9 Limestone purity classification according to [9,12]

Location (1)							
Parameters	SL1,1	SL1,2	SL1,3	SL1,4	SL1,5	Range	Average
CaO%	55.92	54.80	54.51	55.01	55.72	54.51-55.92	55.19
$CaCO_3$ wt.%	98.80	97.85	98.12	98.42	98.82	98.12-98.82	98.40
$MgCO_3$ wt.%	1.09	1.12	1.45	1.02	1.14	1.02-1.45	1.16
Limestone classification	Very high to high purity limestones						
Location (2)							
Parameters	SL2,1	SL2,2	SL2,3	SL2,4	SL2,5	Range	Average
CaO%	54.95	55.80	53.51	54.01	53.72	53.51-55.80	54.40
$CaCO_3$ wt.%	98.23	97.90	98.55	98.51	98.82	97.90-98.82	98.41

$MgCO_3$ wt.%	1.10	1.10	1.49	1.08	1.15	1.10-1.49	1.18
Limestone classification	High purity limestones						
Location (3)							
Parameters	SL3,1	SL3,2	SL3,3	SL3,4	SL3,5	Range	Average
CaO%	55.19	54.95	53.52	54.34	53.90	53.52-55.19	54.38
$CaCO_3$ wt.%	98.43	97.10	98.40	98.35	97.89	97.10-98.43	98.03
$MgCO_3$ wt.%	1.13	1.20	1.40	1.18	1.25	1013-1.25	1.23
Limestone classification	High purity limestones						

7.4. Major Elements Concentration

According to a quantitative chemical study of the key elements, the most common mineral in the limestone samples is calcite. As indicated in Tables 1, 2, and 3, the geochemical analysis of limestone samples from various study area locations showed average CaO contents of 55.19%, 54.40%, and 54.38% for the three locations, respectively. Because the limestone is mostly calcite, the results show that calcium oxide (CaO) is the predominant component of the limestone, indicating that the limestone from this deposit is appropriate for cement production. The typical values of magnesium oxide (MgO) are 0.50%, 0.77%, and 1.07%; the lower value also suggests or supports an entirely calcite process. The percentage of silicon (SiO_2) is quite lower, with an average of 0.018%, 0.02% and 0.04%. This indicates the existence of non-carbonate silicate minerals like chert, silt and clay-sized sand particles. Fe_2O_3 amounts tend to be very low also, with an average 0.006%, 0.01% and 0.008% for the three locations respectively. This indicates that the depositional environment has a slightly lower oxidation effect. The low values show that the environment that deposits is a reducing type, which means that the pH of the water and the surrounding environment's redox activity do not support the precipitation of ferric iron (Fe^{3+}) to iron ferrous iron (Fe^{2+}), and therefore the oxides are leached away [14].

Figures 7 through 10 of selected major oxides of limestones are plotted against CaO% such as Al_2O_3 %, SiO_2%, MgO% and Fe_2O_3%. Both Al_2O_3 and SiO_2 exhibit weak negative correlations between the two variables, while MgO and Fe_2O_3 show fair negative correlations.

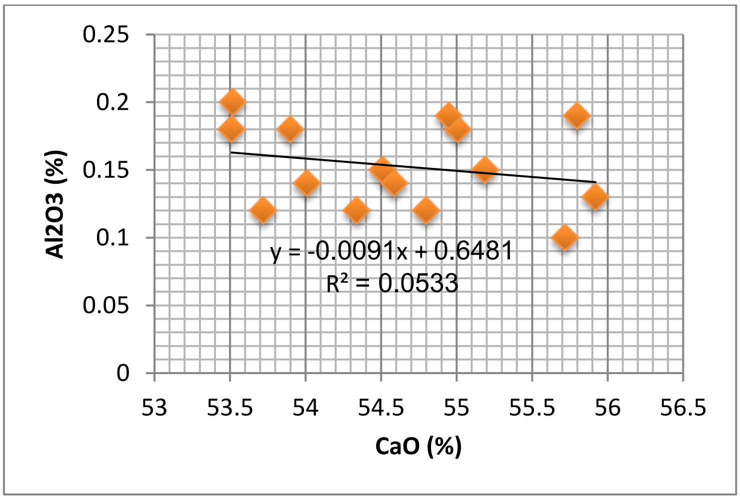


Figure 8: Cross plotting between CaO and Al_2O_3 oxides for limestone samples

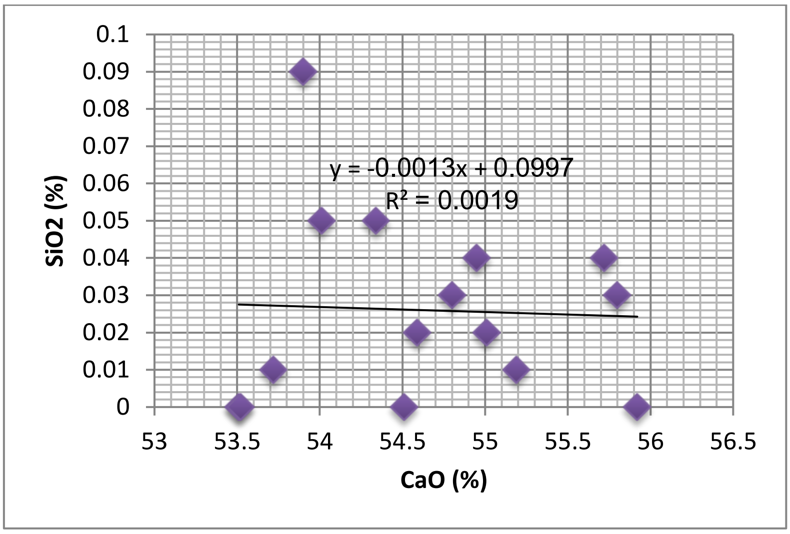


Figure 9: Cross plotting between CaO and SiO_2 oxides for limestone samples

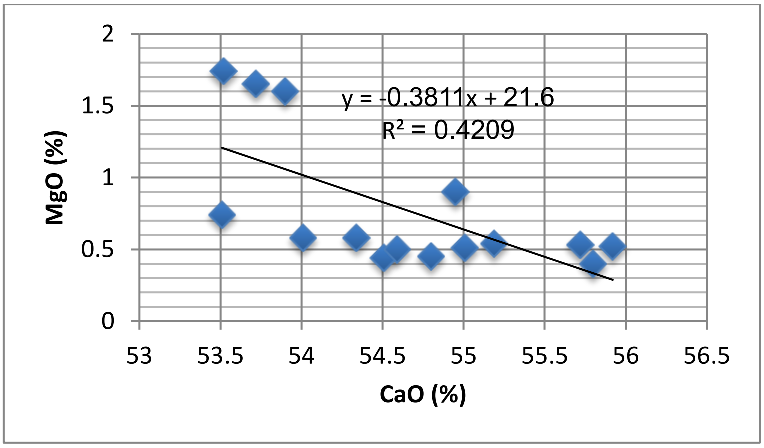


Figure 10: Cross plotting between CaO and MgO oxides for limestone samples

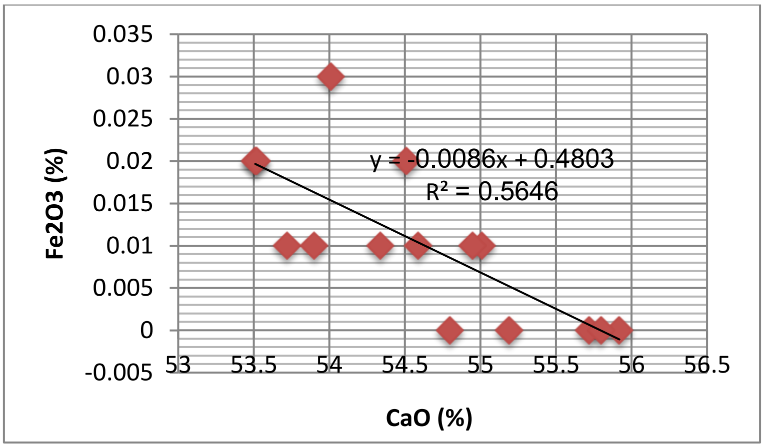
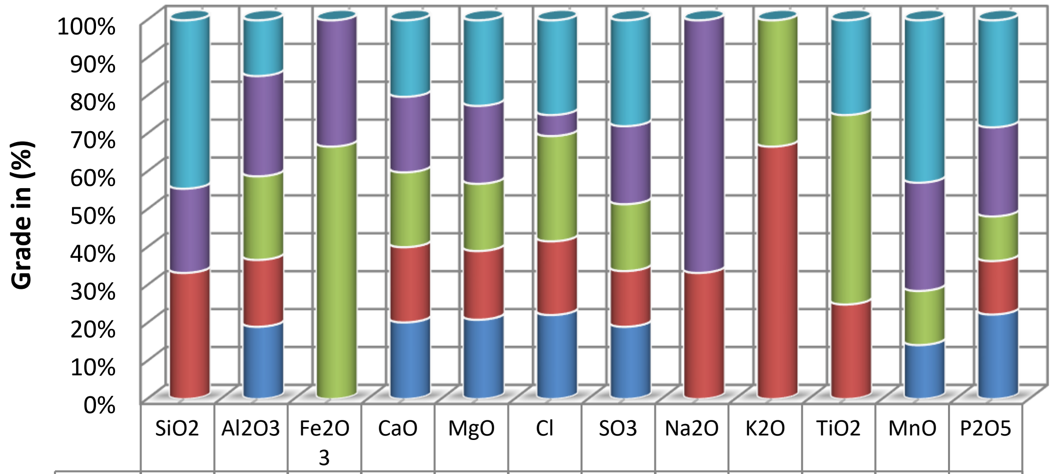


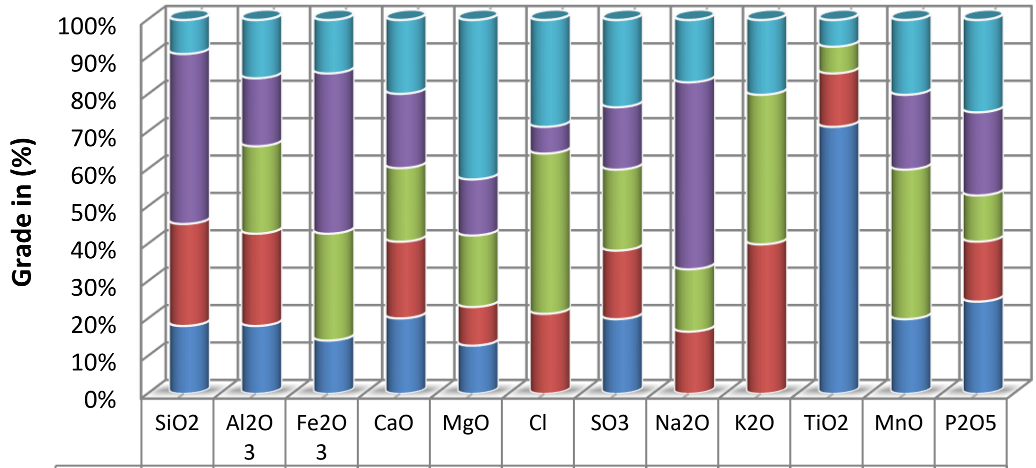
Figure 11: Cross plotting between CaO and Fe_2O_3 oxides for limestone samples

Conversely, Figures 11 through 14 graphically depict the principal oxide distribution in site samples for raw materials (clay and limestone). The environment of deposition, which is represented in the chemical composition of these carbonate rocks, may be responsible for the grading in proportion of these oxides showing some variation from one place to another.



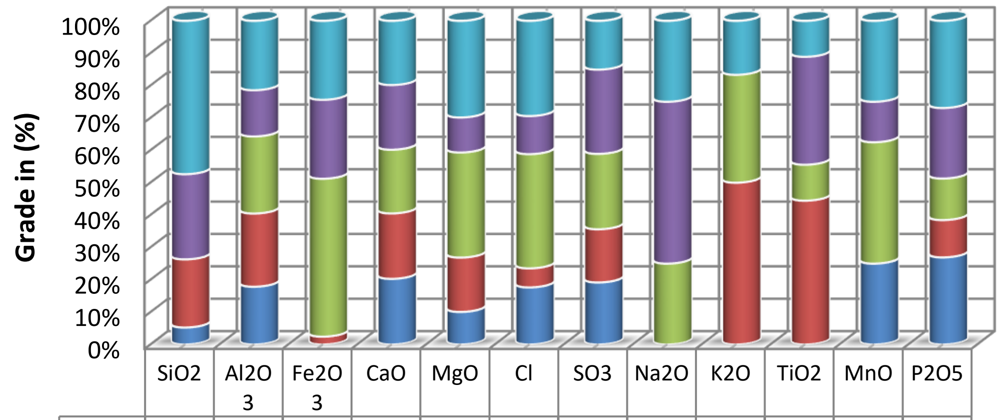
	SiO2	Al2O3	Fe2O3	CaO	MgO	Cl	SO3	Na2O	K2O	TiO2	MnO	P2O5
SL1,5	0.04	0.1	0	55.72	0.56	0.09	0.19	0	0	0.001	0.03	0.24
SL1,4	0.02	0.18	0.01	55.01	0.51	0.02	0.14	0.002	0	0	0.02	0.2
SL1,3	0	0.15	0.02	54.51	0.44	0.1	0.12	0	0.001	0.002	0.01	0.1
SL1,2	0.03	0.12	0	54.8	0.45	0.07	0.1	0.001	0.002	0.001	0	0.12
SL1,1	0	0.13	0	55.92	0.52	0.08	0.13	0	0	0	0.01	0.19

Figure 12: Major oxide concentrations in limestone rocks for location (1)



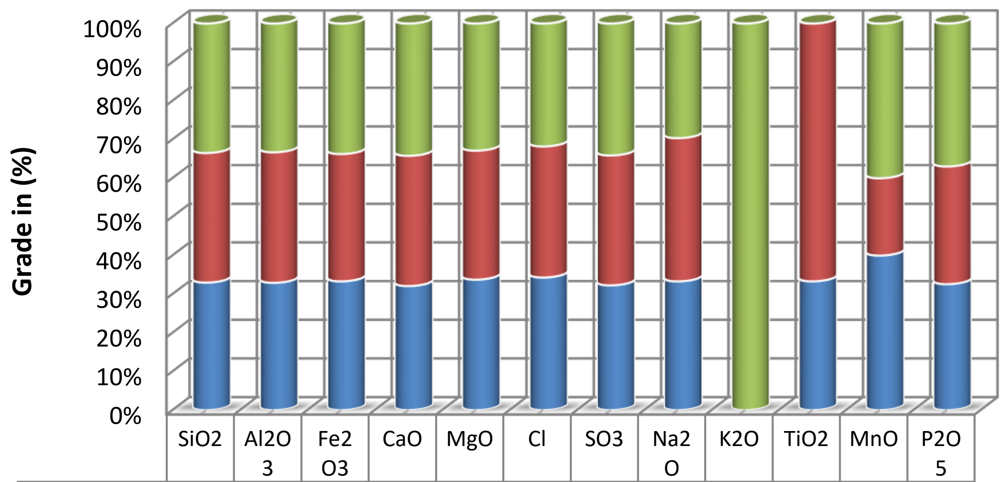
	SiO2	Al2O3	Fe2O3	CaO	MgO	Cl	SO3	Na2O	K2O	TiO2	MnO	P2O5
SL2,5	0.01	0.12	0.01	53.72	1.65	0.08	0.14	0.001	0.001	0.001	0.01	0.2
SL2,4	0.05	0.14	0.03	54.01	0.58	0.02	0.1	0.003	0	0	0.01	0.18
SL2,3	0	0.18	0.02	53.51	0.74	0.12	0.13	0.001	0.002	0.001	0.02	0.1
SL2,2	0.03	0.19	0	55.8	0.4	0.06	0.11	0.001	0.002	0.002	0	0.13
SL2,1	0.02	0.14	0.01	54.95	0.5	0	0.12	0	0	0.01	0.01	0.2

Figure 13: Major oxide concentrations in limestone rocks for location (2)



	SiO2	Al2O3	Fe2O3	CaO	MgO	Cl	SO3	Na2O	K2O	TiO2	MnO	P2O5
SL3,5	0.09	0.18	0.01	53.9	1.6	0.1	0.11	0.001	0.001	0.001	0.02	0.21
SL3,4	0.05	0.12	0.01	54.34	0.58	0.04	0.19	0.002	0	0.003	0.01	0.17
SL3,3	0	0.2	0.02	53.52	1.74	0.12	0.17	0.001	0.002	0.001	0.03	0.1
SL3,2	0.04	0.19	0.001	54.95	0.9	0.02	0.12	0	0.003	0.004	0	0.09
SL3,1	0.01	0.15	0	55.19	0.54	0.06	0.14	0	0	0	0.02	0.21

Figure 14: Major oxide concentrations in limestone rocks for location (3)



	SiO2	Al2O3	Fe2O3	CaO	MgO	Cl	SO3	Na2O	K2O	TiO2	MnO	P2O5
Location (3)	37.06	7.98	4.15	21.4	3.25	0.75	1.32	0.24	0.01	0	0.02	0.17
Location (2)	37.1	8.09	4.05	21.08	3.3	0.8	1.3	0.3	0	0.02	0.01	0.14
Location (1)	36.55	7.9	4.1	20.1	3.34	0.81	1.25	0.27	0	0.01	0.02	0.15

Figure 15: Major oxide concentrations in clay rocks for the three locations

7.5. Ratio Factors Analysis

Cement making requires essential major oxides in the raw material for balancing and proper burning for a good-quality cement product. Major oxides like CaO, SiO_2, Al_2O_3, and Fe_2O_3 require various confirmation tests, such as lime saturation factor (LSF), silica modulus (SM), and alumina modulus (AM) [15].

The ratio factors analyses have been calculated by applying the various equations of moduli as shown in Table 10.

Table 10 Summarizing the applied equations of different factors and moduli

Modulus	Equations [16]
Lime Saturation Factor	$LSF = \frac{CaO}{2.8(SiO_2)+1.2(Al_2O_3)+0.65(Fe_2O_3)}$ (Eq. 1)
	If MgO less than 2.0% and AM more than 0.64, then $LSF = \frac{CaO}{2.80(SiO_2)+1.65(Al_2O_3)+0.35(Fe_2O_3)}$ (Eq. 2)
	If AM less than 0.64, then $LSF = \frac{CaO}{2.80(SiO_2)+1.65(Al_2O_3)+0.35(Fe_2O_3)}$ (Eq. 3)
Lime Standard	$LSt = \frac{100\ CaO}{2.8(SiO_2)+1.18(Al_2O_3)+0.65(Fe_2O_3)}$ (Eq. 4)
Silica Modulus	$SM = \frac{SiO_2}{Al_2O_3+Fe_2O_3}$ (Eq. 5)
Hydraulic Modulus	$HM = \frac{CaO}{SiO_2+Al_2O_3+Fe_2O_3}$ (Eq. 6)
Alumina Modulus	$AM = \frac{Al_2O_3}{Fe_2O_3}$ (Eq. 7)
Liquid phase (%)	$Lf = \frac{71.0}{0.53+SM}$ (Eq. 8)
Sulphur to Alkali Ratio	$\frac{SO_3}{Alkali} = \frac{SO_3/80}{\left(\frac{K_2O}{94}\right)+\left(\frac{0.5Na_2O}{62}\right)}$ (Eq. 9)
	$\frac{SO_3}{Alkali} = \frac{SO_3/80}{\left(\frac{K_2O}{94}\right)+\left(\frac{Na_2O}{62}\right)}$ (Eq. 10)

7.5.1. Lime Saturation Factor (LSF)

Because it comprises CaO, the main mineral component of cement, the lime saturation factor is essential to the production of cement. It shows that in order to make up for the quantity of silica-alumina and iron oxides in the permissible limestone, additional clay must be added throughout the cement-making process. LSF is around 1.0, meaning that the lime content precisely balances the silica, alumina, and ferric oxide concentrations. Clinker may include free lime if the ratio is greater than 1.0 [17].

The LSF regulates the clinker's alite (tricalcium silicate, C3S) to belite (dicalcium silicate, C2S) ratio. Alite and belite are more prevalent in high LSF than in clinker. Kiln feed control is the primary use for the LSF [18]. The excess free lime has nothing to do with and will continue to be free lime if the LSF is greater than 1.0 [19]. In reality, raw material mixing is never flawless, and there are always areas of the clinker where the LSF is significantly lower than the overall objective. Free lime is probably present in the clinker if the value is greater than 1.0. The average LSF value in the current limestone samples is 0.177, which is less than the 1.0 value given in Tables 12, 13, and 14. It shows that the concentration of CaO values in limestones is lower, while the other limestones have a consistent range. A number higher than 1.0 indicates that free lime is most likely present in the clinker. This is because, in theory, all of the free lime ought to have been combined with belite to create alite at LSF = 1. If the LSF value is higher than 1.0, the excess free lime has nothing to do with and will remain free lime [20].

However, the investigated limestone in the three locations have <1.0 values, so these are suitable for this utilization for cement manufacture.

7.5.2. Alumina Modulus (AM)

An essential component of the cement-making process is the alumina ratio. The possible relative ratio of aluminate to the ferrite phase in the clinker is determined by the alumina ratio. An increase in clinker AR indicates that the clinker has less ferrite and more aluminate. There is more alumina and less ferrite when the alumina ratio is greater, and vice versa [18].
Ordinary Portland cement clinker typically has an alumina ratio of 1.0 to 4.0 [18]. The average value of the AR in the current samples is 1.95 (Tables 12, 13, & 14). The majority of samples will have an alumina modulus between 1.92 and 1.99.

7.5.3. Silica Modulus (SM)

In the production of cement, the silica ratio is crucial and has a big impact on the combustion process [18]. The quantity of liquid phase decreases as the silica ratio rises, and vice versa. Thus, the development of the liquid phase is significantly influenced by the silica ratio. The silica ratio was calculated using the following formula.The increased silica concentration results in poor coating properties and combustion challenges. The kiln needs greater heat to operate as the silica ratio rises [21]. Cement will develop ring formation if the silica ratio is less than the recommended ratio. The silica ratio varies from 1.9 to 3.2, per [22]. Poor consistency in the kiln feed may be indicated by a significant fluctuation in the silica ratio in the clinker. According to Tables 12, 13, and 14, the average silica ratios for the limestone samples used in this investigation are 0.25, 0.18, and 0.35 at locations 1, 2, and 3, respectively. This suggests that the samples have a standard specified limit of SiO_2 content and are taken into consideration as a limestone resource for cement production. Since this limestone is nearly inside the usual ratio, there won't be any grinding issues, therefore think about producing regular Portland cement (OPC). The silica ratio and its distributions for various locales show some variance.

7.6. Quality Control Coefficients of Raw Materials

The assessment of raw materials of this study based on the calculations of apparent values of clinker constituents such as C2S, C3S, C3A and C4AF using Bogue's formulae, that rely on the typical chemical composition of limestone rocks suggested by Klieger [23], Bayles [24] and Moore [25]. The equations are expressed as following:

$$C3S = 4.071(CaO) - 7.602(SiO_2) - 4.479(Al_2O_3) - 2.859(Fe_2O_3) - 2.852(SO_3) \quad (11)$$

$$C2S = 2.867\,(SiO_2) - 0.7544(C3S) \quad (12)$$

$$C4AF+C2F = 2.1(Al_2O_3) + 1.702(Fe_2O_3) \quad (13)$$

Table 11 gives calculations results, whereas, these calculations of these parameters revealed that there a variance of these values for the studied locations, and this owing to the different of chemical composition of raw materials in the investigated samples, and within the range of requirements as raw materials for cement industry.

Table 11 Typical chemical composition of limestone and Bogue apparent values

Limestone types / Components	A	B	C	D	E
SiO_2	4.00	13.60	2.00	12.05	2.96
Al_2O_3	0.77	2.50	0.80	3.19	0.79
Fe_2O_3	0.30	0.90	0.20	1.22	0.30
CaO	51.4	43.4	52.90	43.50	52.30
MgO	1.30	3.20	0.90	1.68	1.30
SO_3	0.10	0.10	0.20	0.56	0.03

LOI	42.0	35.6	42.50	36.21	42.05
Na_2O	0.01	-	-	0.12	0.04
K_2O	0.02	0.60	0.20	0.72	0.20
∑	99.90	99.60	99.70	99.25	99.97
Apparent values					
C3S	173.0	55.0	155.9	60.7	184.6
C2S	-119.0	-2.5	-111.9	-11.30	-130.80
C4AF+C2F	0.9	2.70	0.60	3.70	0.90
References	Moore 1996	Bayles 1985	Bayles 1985	Klieger 1985	Klieger 1985

Table 12 Geochemical modulus and quality control coefficients for limestones of location (1)

Location (1)							
Ratios and quality coefficients	SL1,1	SL1,2	SL1,3	SL1,4	SL1,5	Range	Average
Silica Modulus (SM)	--	0.250	--	0.105	0.40	0.40-0.250	0.25
Hydraulic Modulus (HM)	430.15	365.33	320.65	261.95	398.0	261.95-430.15	355.22
Alumina Modulus (AM)	--	--	7.50	0.18	--	0.18-7.50	3.84
Lime Saturation Factor (LSF)	260.70	194.33	214.30	154.31	201.16	154.31-260.70	204.96
Liquid phase (%)	133.96	91.03	133.96	111.81	76.34	76.34-133.96	109.42
C3S	226.83	222.04	220.84	225.45	225.54	220.84-226.83	224.14
C2S	-171.12	-167.42	-166.60	-170.02	-169.94	-166.60-171.12	-169.02
C4AF+C2F	0.273	0.252	0.348	0.395	0.210	0.210-0.395	0.295

Table 13 Geochemical modulus and quality control coefficients for limestones of location (2)

Location (2)							
Ratios and quality coefficients	**SL2,1**	**SL2,2**	**SL2,3**	**SL2,4**	**SL2,5**	**Range**	**Average**
Silica Modulus (SM)	0.133	0.158	--	0.294	0.077	0.077-0.294	0.176
Hydraulic Modulus (HM)	323.24	253.64	267.55	245.50	383.71	245.50-383.71	294.73
Alumina Modulus (AM)	14.0	--	9.0	4.67	12.0	4.67-14.0	8.6
Lime Saturation Factor (LSF)	189.17	140.38	176.02	141.57	234.07	140.38-234.07	176.24
Liquid phase (%)	121.78	103.20	133.96	86.17	116.97	86.17-133.96	112.416

C3S	222.55	225.77	216.61	218.50	217.65	216.61-225.77	220.22
C2S	-167.83	-170.24	-163.41	-164.69	-164.17	-163.41-170.24	-166.07
C4AF+C2F	0.311	0.399	0.412	0.345	0.269	0.269-0.412	0.347

Table 14 Geochemical modulus and quality control coefficients for limestones of location (3)

Location (3)							
Ratios and quality coefficients	SL3,1	SL3,2	SL3,3	SL3,4	SL3,5	Range	Average
Silica Modulus (SM)	0.067	0.209	--	0.358	0.474	0.067-0.474	0.347
Hydraulic Modulus (HM)	344.94	237.88	243.27	301.89	192.50	192.50-344.94	264.09
Alumina Modulus (AM)	--	9.0	10.0	12.0	18.0	9.0-18.0	10.0
Lime Saturation Factor (LSF)	200.33	129.04	158.81	159.12	97.56	97.56-200.23	148.97
Liquid phase (%)	118.93	96.08	133.96	79.96	70.72	70.72-133.96	99.93
C3S	223.53	222.26	216.44	219.73	217.59	216.44-223.53	219.91
C2S	-168.60	-167.56	-163.28	-165.62	-163.90	-163.28-168.60	-165.79
C4AF+C2F	0.315	0.401	0.454	0.269	0.395	0.269-0.454	0.366

As seen in Figures 15, 16, and 17, the computed different moduli and quality control coefficients of the examined limestone rock samples in the three sites under study were visually depicted on a radar diagram. The chemical composition of each form of rock, which is mostly dependent on the nature of depositional under specific geological circumstances, is often responsible for the variances observed in these plots across rock types and places.

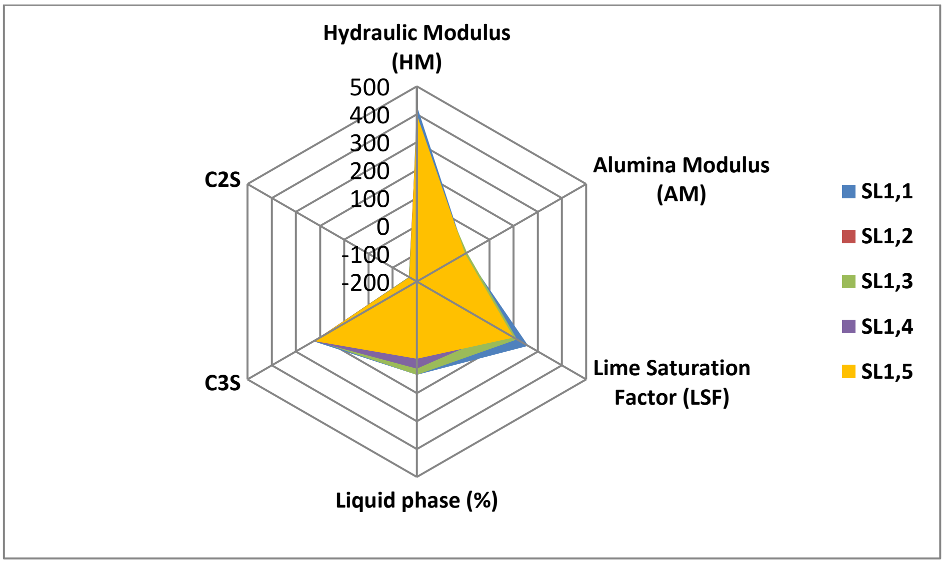


Figure 16: Radar diagram showing various parameters of limestone samples for location (1)

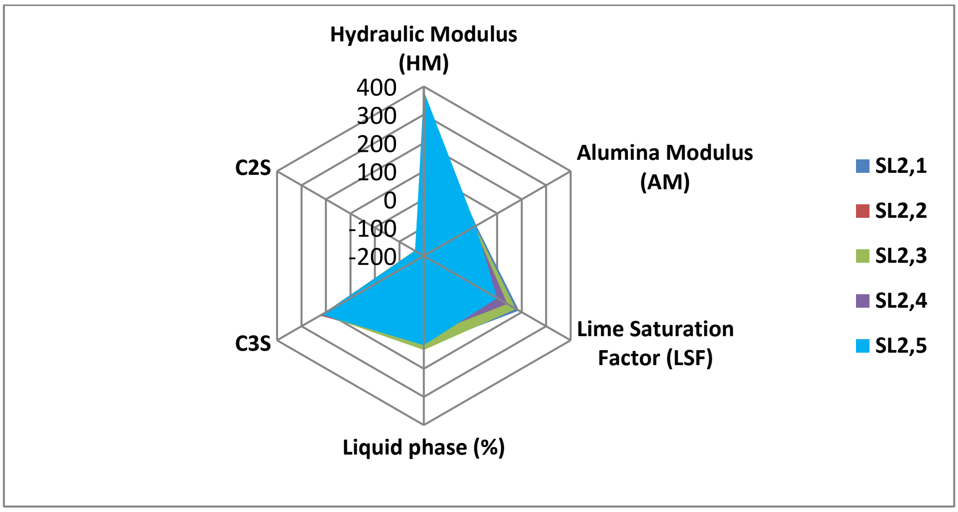


Figure 17: Radar diagram showing various parameters of limestone samples for location (2)

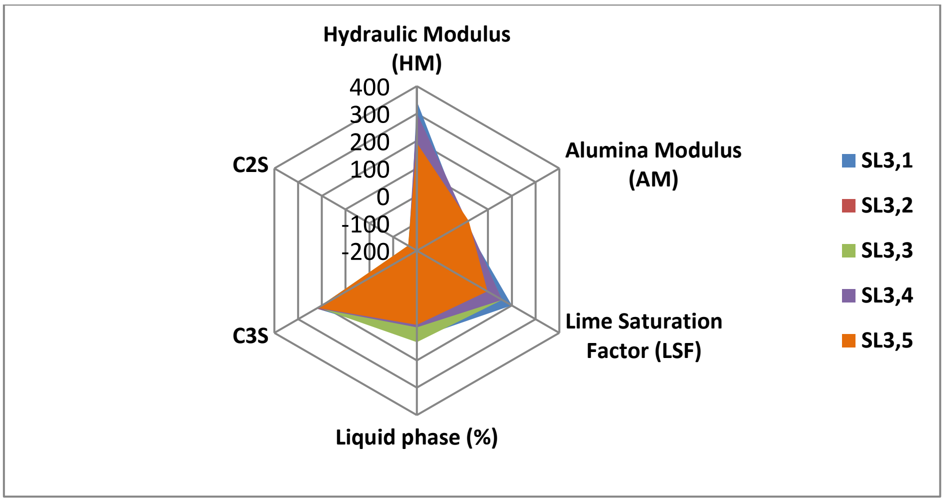


Figure 18: Radar diagram showing various parameters of limestone samples for location (3)

The comparison between Bogue apparent values and the calculated values of studied limestone of the investigates areas for parameters C2S and C3S show a little variance through the investigated parameters of Klieger [23], Bayles [24] and Moore [25], as well as for the examined rocks (Figure 19).

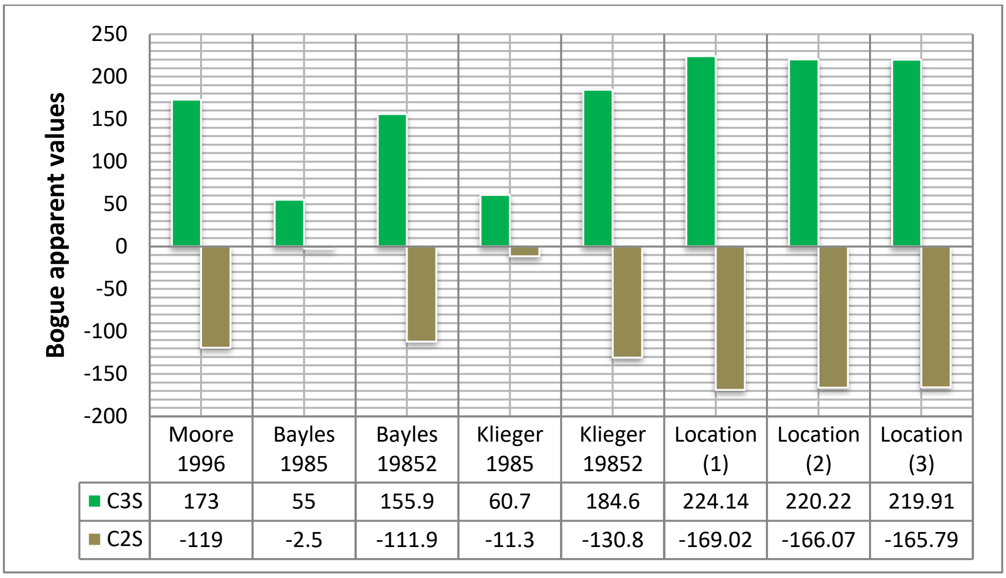


Figure 19: Comparison between Bogue C2S, C3S and studied limestone locations

7.8. Geochemical Suitability of Limestone

This work investigated limestone samples in the studied area aiming to distinguish the potentiality of utilizing the rocks as a natural source of raw materials for cement manufacturing.

Location (1)

In Table 12, the all-major oxide elemental averages concentration ratios display little chemical variation observed in this area: CaO (55.19%), SiO_2 (0.018%), Al_2O_3 (0.14%), Fe_2O_3 (0.006%), MgO (0.50%), and LOI (43.59%). This limestone is rich in CaO% and poor in MgO% and nearly zero SiO_2%. SiO_2 has a relatively low and variable concentration in this limestone. The ratio factor analysis of lime saturation factor, alumina modulus, and silica modulus is in a closely acceptable range. Hence, this limestone was most suitable for cement manufacture according to the standard specifications.

Location (2)

Major oxide elemental averages concentrations in this location are provided in Table 13, and this limestone is homogeneous in chemical composition. From the result, the chemical variation observed in this location was CaO (54.40%), SiO_2 (0.02%), Al_2O_3 (0.15%), Fe_2O_3 (0.01%), MgO (0.77%), and LOI (43.97%). This limestone is rich in CaO% and poor in MgO% and SiO_2%. The ratio factor analysis results confirm that the lime saturation factor is within the range, and the alumina modulus and silica modulus are below the acceptable range. Hence, this limestone was suitable for cement manufacture according to the standard specifications.

Location (3)

These limestones are categorized as highly fossiliferous and brown to cream-coloured. The results of the analyses were presented in Table 14, and no wide chemical variation is observed in this location in CaO (55.38%), SiO_2 (0.04%), Al_2O_3 (0.17%), Fe_2O_3 (0.008%), MgO (1.07%), and LOI (43.71%). This limestone is also suitable for limestone due to its high CaO%. The ratio factor analysis results highlight that the lime saturation factor and alumina modulus are within the range, while the silica modulus is below the acceptable range. Hence, this limestone was suitable for cement manufacture according to the standard specifications after some clay adjustments.

7.9. Blending of Raw Materials

The material composition of Portland cement is usually expressed by the following indices: hydraulic modulus (HM), silica modulus (SM) and alumina (Iron) modulus (AM).

In addition, as a reference of limestone content, limestone reference "K" (indicated in %, lime saturation degree L.S.D. or lime saturation Factor LSF) is used which is 90 to 95 for ordinary Portland cement.

Provided that the blending ratio of lime materials is given, the amount of materials necessary to produce 1 kg of clinker is expressed as follows: when limestone is decomposed by heating, 44% carbon oxide is released as shown in the following equation, resulting in weight reduction:

$$\begin{array}{ccccc} Ca\ C\ O_3 & = & Ca\ O & & C\ O_2 \\ (40+12+48) & = & (40+16) & + & (12+32) \\ 100\ CaCO_3 & = & 56\ CaO & + & 44CO_2 \end{array}$$

Approximately 7% water of crystallization is taken out of the clay. Therefore, the ratio of the amount of the required blending material and the clinker to be obtained can be calculated by the following formulas:

$$a = 1 - \frac{0.44 \times \%CaCO_3}{100} - \frac{0.07 \times (100 - \%CaCO_3)}{100} = \frac{Clinker\ kg}{Blendig\ material\ kg} \qquad (14)$$

$$b = \frac{1}{a} = \frac{Blendig\ material\ kg}{Clinker\ kg} \qquad (15)$$

$$c = \frac{\%CaCO_3 \times 56}{a \times 100} = CaO\ percentage\ in\ clinker \qquad (16)$$

For example, ignition loss when the blending material including 80% limestone is heated can be obtained as follows:

From $CaCO_3$ 0.9840 × 0.44 kg CO_2 = 0.433 kg CO_2 [Location (1)]
From Clay 0.016 × 0.07 kg H_2O = 0.001 kg H_2O

Total ignition loss = 0.434 kg

From $CaCO_3$ 0.9841 × 0.44 kg CO_2 = 0.433 kg CO_2 [Location (2)]
From Clay 0.016 × 0.07 kg H_2O = 0.001 kg H_2O

Total ignition loss = 0.434 kg

From $CaCO_3$ 0.9803 × 0.44 kg CO_2 = 0.431 kg CO_2 [Location (3)]
From Clay 0.020 × 0.07 kg H_2O = 0.001 kg H_2O

Total ignition loss = 0.432 kg

This means that about 0.567 kg, 0.569kg and 0.568 kg of clinker for locations (1), (2) and (3) respectively, can be produced from 1 kg of blending materials including the $CaCO_3$. In another words, 1.764 kg, 1.757 kg and 1.761 kg of raw materials is theoretically required for producing 1 kg of clinker for each location respectively.

Raw materials actually used are composed of various constituents, which should be skillfully blended to produce such raw materials as will satisfy the values within the index range. If the components thus obtained fail to satisfy the requirement, the third substance will have to be added to fill the insufficiency.

1. When the component ratio of $CaCO_3$ in the blending material is given:

The blending ratio of two raw material components can easily be obtained by applying a proper value to X.

Supposing that X $CaCO_3$ content required in the material mixture is 80%, a formula can be obtained based on the following material content, as given below:

For location (1):

$$a = CaCO_3 contained\ in\ limestone = \frac{55.19 \times 100}{56} = 98.55\%$$

$$b = CaCO_3 contained\ in\ clay = \frac{20.10 \times 100}{56} = 35.89\%$$

For location (2):

$$a = CaCO_3 contained\ in\ limestone = \frac{54.40 \times 100}{56} = 97.14\%$$

$$b = CaCO_3 contained\ in\ clay = \frac{21.08 \times 100}{56} = 37.64\%$$

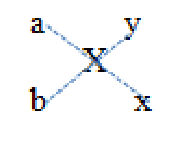
For location (3):

$$a = CaCO_3 contained\ in\ limestone = \frac{54.38 \times 100}{56} = 97.11\%$$

$$b = CaCO_3 contained\ in\ clay = \frac{21.40 \times 100}{56} = 38.22\%$$

The data can be tabulated as following:

Locations	x values	y values
Location (1)	x = 98.55 – 80 =18.55	y = 80 – 35.89 = 44.11
Location (2)	x = 97.14 – 80 = 17.14	y = 80 – 37.64 = 42.36
Location (3)	x = 97.11 – 80 = 17.11	y = 80 – 38.22 = 41.78



Blending ratio for location (1) = $\frac{Limestone}{Clay} = \frac{44.11}{18.55} = \frac{2.38}{1}$

Blending ratio for location (2) = $\frac{Limestone}{Clay} = \frac{42.36}{17.14} = \frac{2.47}{1}$

Blending ratio for location (3) = $\frac{Limestone}{Clay} = \frac{41.78}{17.11} = \frac{2.44}{1}$

8. CONCLUSION

Based on the previous findings the following conclusions can be drawn:

1. The studied limestones show variable lithology e. g. white limestone, white yellowish limestone and fossiliferous limestone, depending on chemical composition and depositional environments.
2. The geochemical classification according to Al_2O_3–CaO–(MgO+FeO_t) ternary diagram and Ca/Mg ratio, limestones are classified as pure limestones and fall within the standard used for cement industry.
3. Based on the obtained chemical analyses and according to CaO% in addition to the $CaCO_3$%, limestones classified as high purity limestones and very high purity limestones

4. Ratio factors analysis using lime saturation factor (LSF), silica modulus (SM), and alumina modulus (AM) proved that these rocks within the specifications of these factors.
5. The calculated various moduli and quality control coefficients such as C2S, C3S, C3A and C4AF using Bogue's formulae of limestone rock samples reflected that there some variations between rock types and locations, this is usually can be attributed to the chemical composition for each type which is primarily relay on the nature depositional under a certain geological conditions
6. The geochemical suitability of limestone proved that these raw materials are suitable for cement manufacture according to the standard specifications.
7. Blending ratios of raw materials for both limestone and clay rock samples show little variation between the locations (1), (2) and (3) whereas, 1:2.38, 1:2.47 and 1:2.44 respectively.
8. The present work is helpful for natural resources as a raw materials and are most suitable for cement manufacturing to achieving the sustainable development for future country economic development.

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Compliance with ethical standards

Disclosure of conflict of interest

The authors declare that they have no conflict of interest.