

# Physiochemical and Mineralogical Investigations of Some Raw Materials in Wadi Abou Al Gomel Area, Eastern Region, Libya

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# المخلص:

أجريت هذه الدراسة في منطقة وادي أبو القمل من خلال اختيار ثلاثة مواقع هي (1) و (2) و (3) بشرق ليبيا على ساحل البحر الأبيض المتوسط بين 03'00 N ° 32N32° 30'08 و 30'08 و 30'08 و 30'08 ° 00'52° . وتتكون المنطقة من تشكيلتين رئيسيتين هما الخويمات والفائدية اللذان ينتميان إلى العصر الثالث. تتكون الصخور الحجرية بشكل أساسي من الحجر الجيري والدولوميت والحجر الجيري مارلي والصخور الجيرية التي يمثلها الصخر الزيتي. الهدف من هذه الدراسة هو تسليط الضوء على الصخور فيزيائياً وكيميائياً ومعدنيا بالإضافة إلى تقييمها كمواد خام لتصنيع الأسمنت. أظهرت النتائج الكيميائية أن الأكاسيد الرئيسية تظهر تبايئاً في التوزيع والأكسيد الرئيسي هو CaO لتصل إلى 55.15٪ والتي يمكن أن تلبي متطلبات صناعة الأسمنت. وفقًا للخصائص الفيزيائية للعينات المدروسة ، يمكن تصنيفها إلى فتتين هما الصخور الصلبة والصخور شديدة الصلابة. كشفت التحقيقات المعدنية أن صخور الكربونات تنشأ في بيئات مختلفة ، وعادة ما يحدث الأراجونيت والكالسيت عالي المغنيسيوم في البيئات البحرية الضحلة الدافئة عن طريق الترسيب المباشر من مياه البحر أو من الهياكل العظمية لكائنات مختلفة. يشير الدولوميت بين المعادن السائبة في تتابع أوليجوسين-ميوسين إلى تباين في نظام الترسيب من تجاوز الجرف البحري الضحل إلى ارتداد طفيف في من منطقة المد. تراوحت نسب الجبس بين 2 و 5٪. بينما توجد الهاليت بكميات قليلة تراوحت من 1 إلى 4٪ في كل من تشكيلات منطقة المد. تراوحت نسب الجبس بيئة المد والجزر التي تتميز بالتبخر.

الكلمات المفتاحية: المواد الخام، الخواص الفيزيوكيميائية، الخواص المعدنية، التكوينات الصخرية، البيئية الترسيبية.

#### **Abstract**

This study was conducted on Wadi Abou Al Gomel area through three selected sites, namely: (1), (2) and (3), in the east of Libya along the coast Mediterranean Sea, between N32°03'00 N32°08'30 and E23°45' 00 E23°52'00. The area comprises two main formations which are Al Khowaymat and Al Faidiyah that belonging to Tertiary age. The lithology of rocks is mainly limestone, dolomite and marly limestone and argillaceous rocks represented by shale. The objective of this study is to highlight rocks physically, chemically and minerlogically as well as the assessment as raw materials for cement manufacturing. The chemical results revealed that the major oxides exhibit a variation of distribution and the major oxide is CaO to reach 55.15% which meets the requirements of cement industry. According to the physical properties of the studied samples, they can be classified into two categories which are hard rocks and very hard rocks. The mineralogical investigations revealed that the carbonate rocks arise in different environments, aragonite and high-magnesium calcite usually occur in the warm, shallow marine environments by direct precipitation from seawater or from skeletons of various organisms. Dolomite among the bulk minerals in the Oligocene-Miocene successions indicates variation in the sedimentation regime from shallow marine shelf transgression to slightly regressive intertidal. Gypsum percentages ranged between 2 and 5%, while halite occurred in low amounts

varied from 1 to 4% in both Al Khowaymat and Al Faidiyah formations. This reflects supratidal to intertidal environment marked by evaporation.

**Keywords**: Raw materials, Physiochemical properties, mineralogical properties, rock formations, depositional environment.

#### 1. Introduction

In general, the characterization lithology of the studied area is encountered in a few rock types that are represented by calcareous fossiliferous rocks, shale, sandy shale and conglomerate. The stratigraphic succession shows some variation from wadi to another in thickness and extent due the variation of depositional factors and the tectonic movements. The thick sedimentary section exposed in Wadi Abou Al Gomel area, especially noticed along the cliffs overlooking the Mediterranean Sea are classified into two main stratigraphic formations belonging to Early Tertiary. Besides, the five quaternary deposits were recognized near the foot slopes of the cliffs and inward covering older units.

- 1. **Al Khowymat Formation**: dolomitic limestone, yellowish white, hard compact, fossiliferous including; *Globgerina* spp., *Globorotalia* spp. and *Nummulites*.
- 2. **Al Faidiyah Formation**: limestone, faint brown to dark yellow, sandy and marly. Lithologically the formation is made of alternating limestone, marly limestone and clay beds. The beds are nearly horizontal, thin to thin-bedded and highly fossiliferous.

#### 2. Location of Study

The studied area lies at the eastern part of Libya on the coast of Mediterranean Sea at about 6 Km in the west of Tobruk city and about 2.5 Km from the coastal road, between N32°03'00 N32°08'30 and E23°45' 00 E23°52'00 (Fig. 1). The area is characterized by its elevation above sea level which is ranging from 80-120 m. The area comprises different wadis that formed due to the tectonic movement, the major one is Wadi Abou Al Gomel. The study concentrated on this wadi due to its location importance and their good exposures of rock formations.

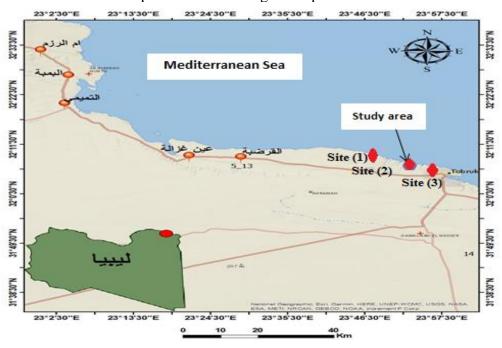


Fig. 1 Sample location map of study area



### 3. Study Objectives

This study was carried out on this area aiming to evaluate some characterizing parameters e. g. physical properties which including specific gravity, water saturation, bulk density and porosity; and chemical properties e.g. the different chemical constituents of the investigated rock samples, as well as the mineralogical composition using XRD analysis technique. On the other hand, the study attempts to assess some rocks e. g. limestone and clay as raw materials for cement manufacturing.

# 4. Literature Review

There are some geological studies that have been conducted at the northeastern region of Libya. The recent studies can be reviewed as follows:

El-Ekhfifi et al. (2017) determined the mineralogical and foraminiferal components of the sand beach of Tobruk, NE Libya. They found that these sediments are consisted of fine to coarse grained sediments rich with benthic foraminifera. Carbonate content is ranging from 60 to 95% while detrital quartz and feldspar varied from 5 to 40% related to erosional factors. They related these components to marine waves, then blown by winds to form exposures of calcarenites around the study area.

Muftah et al. (2017) studied three surface sections; Wadi al Hash and Wadi al Shaigh at Tobruk area and Wadi al Rahib at Al Bardia area. The first location is described as a transgressive event which was recognized at the basal part of the Oligocene - Miocene Al Faidiyah Formation remarking the disconformity surface separating the Oligocene - Miocene Al Faidiyah Formation from the underlying Middle Eocene Darnah Formation. A disconformity surface of a short interval time is traced between the Al Faidiyah Formation and the overlying Al Jaghbub Formation.

Abdulsamad et al. (2018) reported that the Tobruk coastal area of northeastern Cyrenaica (NE Libya) is marked by steep cliffs rising more than 100 m above the Sea and are dissected by wadis coming down from these scarps. The age of these escarpments is of Middle Eocene to Middle Miocene rocks.

Elfeituri et al. (2021) this paper has been carried out to determine the mineralogical and geochemical characteristics of Eocene Darnah limestone which outcrops at Wadi Al Kharsha as an example of the Darnah Formation. The most conspicuous phenomenon is the upward increase of the dolomitization process, whereas, the dolomite content ranges from 4.34 to 15.48 % with an average value of 8.99%. The lime (CaO) represents the major predominant with a high content (48.08%), it can be blend with other carbonates to improve its quality to meet the specifications of raw material for cement manufacture.

# 5. Materials and Methods

The methodology of the study was represented in the repeated field trips, collection of different rock samples that represent the different formations beds. The samples collected from study locations were physically and chemically analyzed for major components at laboratory of Libyan Cement Company at Al Fataih using XRF instrument. In the meanwhile, the mineralogical analysis was performed at AL Mansoura University in Egypt by using XRD technique.

# 6. findings and discussion

The chemical analyses are very important for the assessment of the studied rocks as a raw material that can be used in different applications. Thus the raw materials in this study represented by limestone and clay are chemically evaluated as raw materials for cement

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المجلد 8 – العدد 16 16 Volume 8 – Issue 16

industry. The results of chemical analysis for the studied sites of limestone and clay rocks were presented in Table1

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# **6.1. Chemical Properties**

Chemical properties play an important part in the characteristic of carbonate rocks. Geologists may further use geochemical information to improve understanding of geological processes, including the origin, formation, and composition of rocks and sediments (Rollinson et al., 2014). The concentrations of major oxides have been used to investigate limestone samples of different sites. SiO<sub>2</sub> concentration is very low and sometimes nil. The highest value was recorded in site (3) with an average 4.90%. Also, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> are low concentration, while the other components e. g. Aluminum is mainly hosted in clay minerals and occasionally in detrital feldspars (Albarede, 2009). alkalies (K<sub>2</sub>O & Na<sub>2</sub>O), Mg, TiO<sub>2</sub>, Mn and P<sub>2</sub>O are minor contents (Table 1). CaO is the principal major oxide of most carbonate rocks and its concentration in limestone rock type can be an indication of chemical weathering. The CaO concentration unlike SiO<sub>2</sub> decreases insignificantly as the weathering increase. The highest value of CaO was recorded in site (2) (55.15%).

Table 1 Chemical composition of limestone rocks

			omposition				
	Site (1)		Site (2	2)	Site (3)		
Comp. (%)	Limestone	Clay	Limestone	Clay	Limestone	Clay	
SiO <sub>2</sub>	0.00	12.90	0.00	37.16	0.90	54.95	
Al <sub>2</sub> O <sub>3</sub>	0.166	4.38	0.13	7.99	0.82	6.68	
Fe <sub>2</sub> O <sub>3</sub>	0.020	3.01	0.00	4.15	0.40	3.43	
CaO	54.97	24.96	55.15	21.10	48.60	11.25	
MgO	0.60	14.32	0.54	3.30	3.30 6.91		
Cl	0.053	1.71	0.08	0.80	0.80 0.42		
SO <sub>3</sub>	0.29	0.79	0.14	1.30		0.101	
Na <sub>2</sub> O	0.00	0.37	0.00	0.30	0.05	0.303	
K <sub>2</sub> O	0.00	0.81	0.00	0.00	0.00	1.16	
TiO <sub>2</sub>	0.00	0.42	0.00	0.00	0.03	0.64	
MnO	0.008	0.035	0.01	0.02	0.01	0.046	
P <sub>2</sub> O <sub>5</sub>	0.204	0.130	0.21	0.17	0.20	0.12	
LOI*	43.61	35.95	43.62	23.52	41.2	19.50	
Σ	99.92	99.78	99.88	99.81	99.88	99.96	
CaCO <sub>3</sub>	98.10	44.55	98.43	62.50 86.72		30.78	
MgCO <sub>3</sub>	1.26	29.95	1.13	42.31	6.10	23.58	

 $LOI^* = Loss on ignition$ 

The quantitative identification of these major oxides as shown in Figs. 2 & 3 is important in the characterization of the quality of the examined samples.

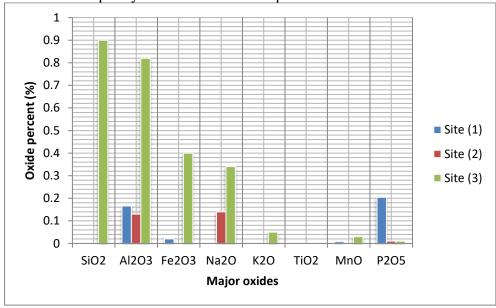


Fig. 2 Major oxides distribution in limestone samples

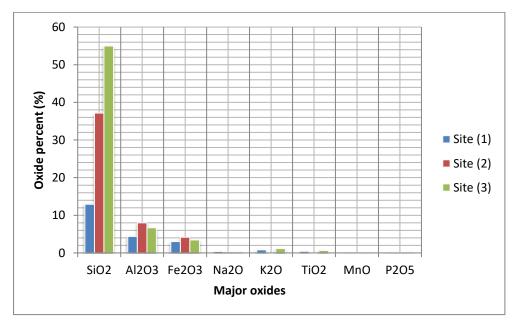


Fig. 3 Major oxides distribution in clay samples

On the other hand, Figs 4 & 5 show the variation of CaO, MgO and LOI in the studied samples, while Figs 6 & 7 depict the amount of content of both CaCO<sub>3</sub> and MgCO<sub>3</sub> in the carbonate samples. It is evident that both CaO and CaCO<sub>3</sub> display a high content to meet the requirement for cement industry specifications as raw material, as well clay rocks.

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Fig. 4CaO and MgO contents and loss on ignition percent (LOI) in limestone samples

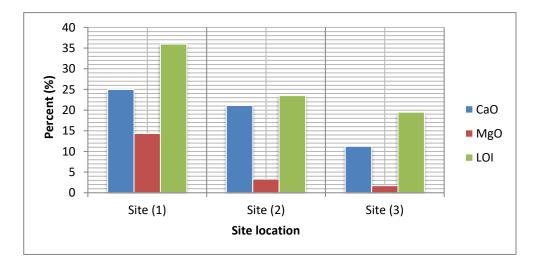


Fig. 5CaO and MgO contents and loss on ignition percent (LOI) in clay samples



Fig. 6 Calcium and magnesium carbonates in limestone samples

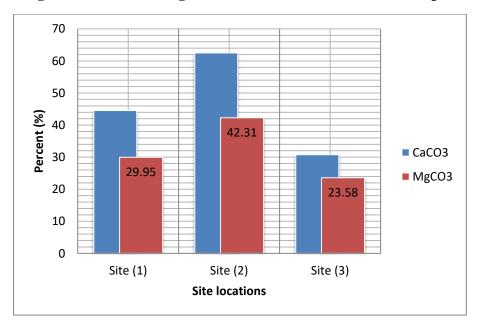


Fig.7 Calcium and magnesium carbonates in clay samples

# **6.2. Physical Properties**

The hand scale analysis showed only a slight difference in color, and the properties e.g. hardness observation highlighted a relevant dissimilarity with respect to the studied rocks. They can be classified into three categories namely, soft rocks, hard rocks and very hard rocks. This may be attributed to dolomitization of limestone. Such difference resulted in one of the main factors controlling the mechanical attitude of the rocks under stress. However, the obtained results of physical properties are presented in Table 2. According to (ASTM C 568-89., 1992). limestone may be classified into three categories. These are, I (Low-Density), for limestone having a density ranging from 1760 through 2160 kg/m³; II (Medium-Density) for limestone having a density greater than 2160 and not greater than 2560 kg/m³ and III (High Density) for limestone

having density greater than 2560 kg/m<sup>3</sup>. At least one type of each category was chosen to be included among the tested specimens. Consequently the major of the investigated limestone samples within the range of category II and III, or in other words can be classified as medium and high density calcareous rocks.

Table 2 Physical properties of limestone rocks for the studied sites

Property	Specific gravity	Water saturation	Bulk density	Porosity
Sites		(%)	(kg/m <sup>3</sup> )	(%)
Site (1)	2.61	2.45	2680	9.10
Site (2)	2.52	2.51	2544	12.84
Site (3)	2.70	2.49	2650	8.15

From a physical point of view, a similar real density characterizes both site (1) and site (3) (2680 & 2650 kg/m³) respectively. This refers to the alteration of some limestone rocks through dolomitization process. This is the only parameter in common between the two varieties of limestone, underlying their same origin from the AL Faydia Formation. On the other hand, these rock types characterized by low porosity 9.10% and 8.25% respectively (Table 2).

#### 6.3. Correlation between Physical Properties

The mutual correlation between physical properties such as specific gravity, water saturation, bulk density and porosity is a useful procedure to understand the dependence of such variables. Several authors discussed about it, with special reference to the influence of porosity on the physical and mechanical properties of rocks, highlighting its weakening role on UCS (e.g. (Tugrul and Gurpinar., 1997); (Hatzor and Palchik.,1998); (Al Harthi et al.,1999); (Sousa et al.,2005; (Zhu et al.,2010); (Pappalardo et al.,2013); (Pappalardo et al.,2016);(Di Benedetto et al.,2015). Results achieved in this research are consistent with literature data, showing that total porosity holds the almost absolute control on the bulk density of a rock (Table 2).

#### 6.4. Mineralogical Analysis

Mineralogical identification was conducted on the investigated rocks using X-ray diffraction techniques (XRD) with Ni-filter and Cu K $\alpha$  radiation. X-ray diffraction was carried out with Philips (PW-3710) diffractometer with generator (PW-1830), Cu target tube and Ni filter at 40 kV and 30 mA. The scanning range was between  $2\Theta$  3° - 60°at scan speed  $2\Theta$  2min-1. The instrument is computerized to measure the peak diffraction in  $2\Theta^\circ$  and d spacing (Å), the peak intensity (integer I) and (I/I $_\circ$ ). Identification of minerals is based on the detection for the characteristic diffraction peaks using the mineral powder diffraction file. Middle Miocene sediments are characterized by significant amount of fauna remnants. It is assumed that most of the carbonate components in the samples are of biogenic origin, with smaller portion which

could arise by precipitation from water. The following minerals were identified using their characteristic peak diffractions in  $2\Theta^{\circ}$  and d spacing (Å) (Table 3).

Table 3 Significant reflections used for identification of the bulk minerals

Minerals	2 <del>0</del> °	d (Å)	Intensity%	ASTM Card no. Chao (1969)
Calcite	29.4	3.04	100	5-586
	39.34	2029	20	
	43.07	2.1	20	
Dolomite	30.9	2.89	100	11-78
	41.22	2.19	30	
	51.14	1.78	20	
Gypsum	11.7	7.56	100	5-628
	20.8	4.27	50	
	31.16	2.68	100 - 30	
Halite	31.7	2.82	100	5 629
Hante	45.48	1.99	50	5-628
	56.5	1.62	20	
Quartz	26.68	3.34	100	5-490
	20.85	4.26	30	
	50.2	1.8	10	

Ten samples were chosen to represent the carbonate rocks for the three sites from the study area and analyzed by X-ray diffraction to determine the bulk mineralogy. Their X-ray diffraction patterns are shown in Fig. 8 and the relative percentages of the total mineral components are given in Tables4 and 5. The data obtained revealed that calcite is the dominant carbonate mineral in all Oligocene-Miocene formations, while dolomite, gypsum, halite and quartz are uncommon and occurred in low amounts in few numbers of samples (Tables 4 and 5).

Variations in their abundances are represented graphically in bar graphs shown in Fig. 9.Distribution of the mineral components in the Oligocene-Miocene formations are shown in Fig. 9.

# 6.5 Distribution and significances of the bulk minerals

#### 6.5.1 Calcite

Calcite is identified from its 20 at 29.4° as a low Mg-calcite in all the analyzed samples (Fig. 9). It is the only dominant carbonate mineral with percentage fluctuated between 81 and 100% in study area (Table 4). While calcite can arise in different environments, aragonite and highmagnesium calcite usually occur in the warm, shallow marine environments by direct precipitation from seawater or from skeletons of various organisms (Lippmann, 1973) (Table 5).

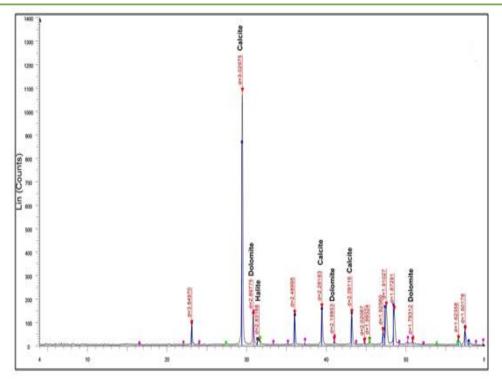
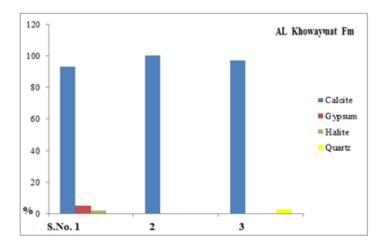


Fig. 8 X-ray diffractograms showing the bulk mineralogical assemblage of the studied samples



Volume 8 – Issue 16 16 16 المجلد 8 – العدد 16

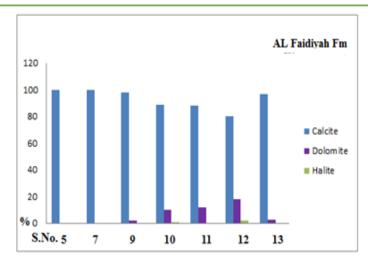
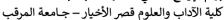


Fig. 9 Histograms showing the relative percentages of the bulk minerals in Al Khowaymat and Al Faidiyah Formations

Table 4 Relative percentages of bulk mineral components (data calculated automatically from X-ray diffraction) for investigated samples.

			Bulk minerals %					
S.	Δ σρ	Formation	Carbonate minerals		Evaporite minerals		Detrital minerals	
No.	D		Calcite	Dolomite	Gypsum	Halite	Quartz	
1	Early	AL	93		5	2		
2	Oligocene	Khowayma t	100					
3			99				1	
5			100					
7			100					
9	Late Oligocene	AL Faidiyah	98	2				
10	- ongocene		89	10		1		
11	Early		89	11				
12	Miocene		81	17		2		
13			97	3				
15			100					





#### 6.5.2 Dolomite

Dolomite is the second carbonate mineral detected from X-ray diffraction analysis. It occurred in low amounts ranged between 2 and 17% in Al Faidiyah Formation rocks in study area (Table 4). Established differences of the mineral content are the result of diagenetic changes caused by the increase of temperature and depth (Grizelj et al., 2011). (Table 5). Recording of dolomite among the bulk minerals in the Oligocene-Miocene successions indicates variation in the sedimentation regime from shallow marine shelf transgression to slightly regressive intertidal.

Table 5 Relative percentages of bulk mineral components (data calculated automatically from X-ray diffraction) in Wadi Abou Al Gomel.

			Bulk minerals %						
S. No.	Age	Formation	Carbonate minerals		Evaporite minerals		Detrital minerals		
			Calcite	Dolomite	Gypsum	Halite	Quartz		
1	Early	AL Khowaymat	94		2	4			
3	Oligocene		90	1		4	5		
8		AL	95			4	1		
9	Late Oligocene-		98			2			
10	Early		99			1			
12	Miocene	Faidiyah	97	3					
13			98	2					

#### 6.5.3 Gypsum and halite

Gypsum and halite recorded only in Al Khowaymat Formation in percentages ranged between 2 and 5%. While halite occurred in low amounts varied from 1 to 4% in both Al Khowaymat and Al Faidiyah formations (Tables 4&5). This reflects supratidal to intertidal environment marked by evaporation.

# **6.5.4 Quartz**

Detrital quartz is the main non-carbonate mineral encountered in very low amounts fluctuated between 1 and 5% in few numbers of samples in Al Khowaymat and Al Faidiyah formations indicating land derived materials from during the regressive phases.

#### 7. Conclusion

In light of the this study the following conclusions can be drawn:

1. The concentration of major oxides have been used to investigate limestone samples of different sites. SiO<sub>2</sub> concentration is very low and sometimes absent . The highest value

recorded in site (3) with an average 4.90%. Also, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> are low concentration, while the other components e. g. alkalies (K<sub>2</sub>O & Na<sub>2</sub>O), Mg, TiO<sub>2</sub>, Mn and P<sub>2</sub>O are minor contents.

- 2. CaO is the principal major oxide of most carbonate rocks and its concentration in limestone rock type can be an indication of chemical weathering. The CaO concentration unlike SiO<sub>2</sub> decreases insignificantly as the weathering increase. The highest value of CaO was recorded in site (2) (55.15%).
- 3. It is evident from the obtained results of chemical analysis that both CaO and CaCO<sub>3</sub> display a high content to meet the requirement for cement industry specifications as raw material, as well clay rocks.
- 4. According to the physical properties of the studied samples, they can be classified into three categories, which are: hard rocks and very hard rocks. This may be attributed to dolomitization of lime stone.
- 5. The mineralogical investigations revealed that the carbonate rocks arise in different environments, aragonite and high-magnesium calcite usually occur in the warm, shallow marine environments by direct precipitation from seawater or from skeletons of various organisms.
- 6. Dolomite among the bulk minerals in the Oligocene-Miocene successions indicates variation in the sedimentation regime from shallow marine shelf transgression to slightly regressive intertidal.
- 7. Gypsum was recorded only in Al Khowaymat Formation in percentages ranged between 2 and 5%. While halite occurred in low amounts varied from 1 to 4% in both Al Khowaymat and Al Faidiyah formations. This reflects supratidal to intertidal environment marked by evaporation.
- 8. Detrital quartz is the main non-carbonate mineral encountered in very low amounts fluctuated between 1 and 5% in Al Khowaymat and Al Faidiyah formations indicating land derived materials from during the regressive phases.

#### 8. Recommendations

In light of this study and the results obtained, the following recommendations can be made:

- ➤ The area will require extensive geological surveys for assessment.
- > General research to be conducted on raw materials that can be used in various industries.
- ➤ Libya has huge reserves of high-quality carbonate rocks, which are used in the cement industry.
- The studied limestone can be used to produce cement.
- > These places could be developed to attract tourists by establishing resorts as part of geotourism.

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