Iraqi Bulletin of Geology and Mining *Vol.16, No.1, 2020* *p 105 – 126*

CHARACTERIZATION AND POTENTIAL INDUSTRIAL UTILIZATION OF THE PERMIAN KAOLIN CLAY DEPOSITS, GA'ARA AREA, WESTERN IRAQ

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Received: 06/ 07/ 2019, Accepted: 05/ 08/ 2019
Keywords: Ball clay; Kaolin clay; Utilization of kaolin clay; Ga'ara kaolin clay; Iraq

ABSTRACT

Representative samples from the investigated kaolin clay at the Ga'ara Depression, Iraqi Western Desert, are assessed in the present work for various industrial applications. Mineralogical, physical and chemical analyses are carried out. The main clay mineral present is kaolinite with occasional traces of illite. The main non-clay admixtures present are quartz, iron oxyhydroxides, anatase and organic matter. Particle size analysis showed that eight deposits are very fine c1ays, three deposits are fine, six deposits are medium and two deposits are coarse clays. The kaolin clays are of various brightness and whiteness, but mostly they are off-white. Ten deposits are plastic and nine of them are super-plastic clays. Silica and alumina are the main constituents. Iron and titanium oxides are present in significant amount whereas the total alkalis are generally low. Nine of the clay deposits are fat, six are moderately fat and four are lean. Although the quality of all white clays can be improved by some processing, nine deposits are sufficiently of high quality to be used after minimal beneficiation in the manufacture of paper, paints, rubber and plastics. In addition, after calcination, some of the samples become suitable for the above end uses. Each of the investigated clay deposit is suitable for use in ceramic manufacture whereas only nine of them are sufficiently high in alumina to be used for the manufacture of chamotte refractories.

تقييم إمكانية الاستخدام الصناعي لرواسب أطيان الكاؤولين البرمية في منطقة الگعرة، غرب العراق

مازن تمراغا، مازن محمد مصطفى و اسماء عبد العزيز ابراهيم

المستخلص

تم في هذه الدراسة تقييم أطيان الكاؤولين في الرواسب المتحرى عنها في منخفض الگعرة الواقع في الصحراء الغربية العراقية للاستعمال كمادة أولية في صناعات مختلفة. بينت فحوصات الأشعة السينية بأن المعدن الطيني الرئيس هو الكاؤولينايت مع آثار للألايت، أما مكونات أخرى مثل معدن الكوارتز وأكاسيد وهيدروكسيدات الحديد والأناتيس. بينت تحليلات المقاسات الحجمية للحبيبات بأن ثمانية من الرواسب تصنف ناعمة جداً وثلاثة منها ناعمة وستة منها متوسطة وأثنتين خشنتين، وبينت مقاسات اللون بأن معظمها بيضاء شاحبة وذات بريق وبياض متباين، كما بينت مقاسات اللدونة بأن

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عشر عينات منها لدنة وتسع فائقة اللدونة. بينت التحليلات الكيميائية بأن السليكا والألومينا هي الأكاسيد السائدة وأن أكاسيد الحديد والتيتانيوم موجودة بكمية لابأس بها بينما مجموع القلويات شحيح. تسع من هذه الرواسب ثرية وستة متوسطة الثراء وأربعة منها هزيلة. بالرغم من إمكانية تحسين نوعية كافة الأطيان البيضاء بإجراء بعض العمليات، فإن تسعة منها عالية المواصفات بما فيه الكفاية للاستخدام، بعد عمليات تهذيب بسيطة، في صناعة الورق والأصباغ والمطاط و\اللدائن، فضلاً عن ذلك تتحسن مواصفات أكثرية الرواسب لهذه الاستخدامات. كما تصلح كافة رواسب الطين التي تم دراستها في الصناعات السيراميكية بينما تصلح تسع منها في صناعة حراريات الشاموط.

INTRODUCTION

A large amount of sedimentary kaolin clay deposits are discovered during the last sixty years in the Ga'ara Depression, Western Desert of Iraq (Fig.l). Prospecting and explorations at the Ga'ara Depression resulted in estimating about 960 million tons of white and coloured kaolin clays (Tamar-Agha *et al*., 2019). Only the Duekhla deposit is currently exploited by open-pit mining. The clay excavated from this mine was mainly consumed in white cement industry (For about two decades) whilst small quantity goes to other industries such as ceramics (wall tiles), paper (as filler) and rubber. Many workers have evaluated the Ga'ara clays especially Duekhla deposit for various industrial uses but mainly ceramics, refractories and paper industries. The major works are those by Mustafa (1981); Jindy and lbrahim (1982); Al-Azzawi and Aly (1983); Hammodi and Abdu11a (1987a); Al-Kaiysi (1989) and Al-Mashaykhi (1991).

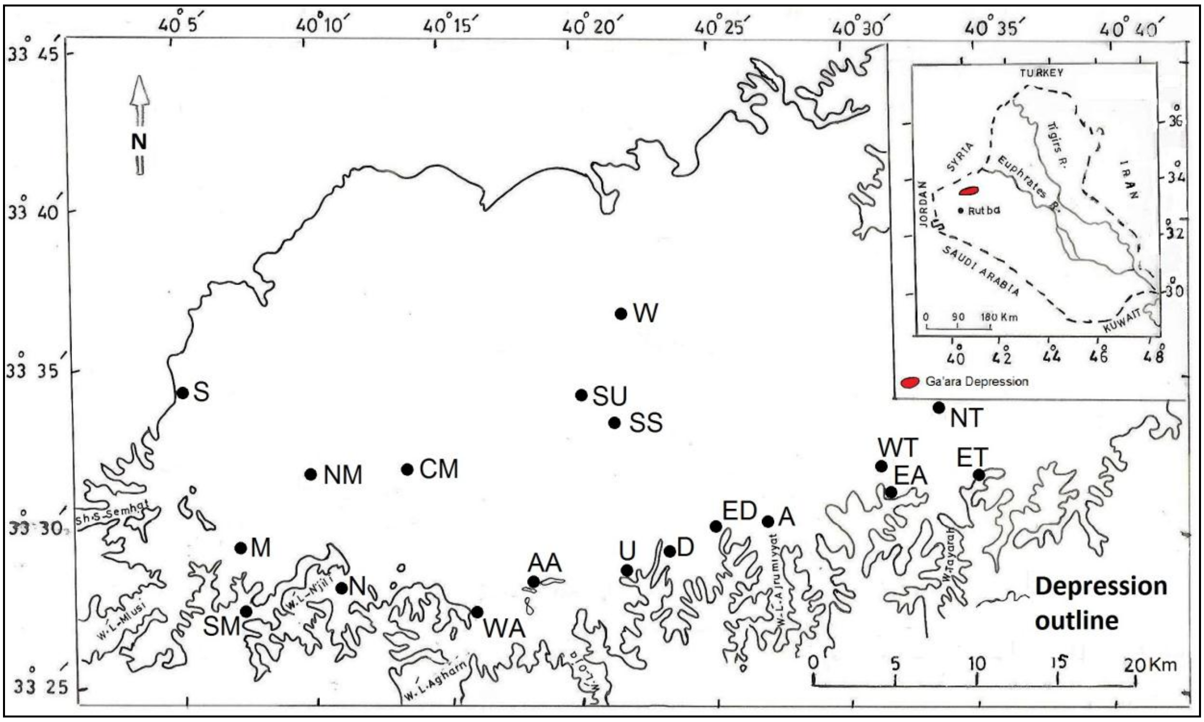

Fig.1: Location map of the investigated kaolin clay deposits at the Ga'ara Depression. NT: North Wadi Tayyarah, ET: East Wadi Tayyarah, EA: East Afaif, WT: West Wadi Tayyarah, A: Tel Afaif, ED: East Duekhla, D: Duekhla, U: Ubairan, AA: Azzlat Al-Agharri, WA: Wadi Al-Agharri, SS: South Ghadir ‘l Sufi, SU: Ghadir ‘l Sufi, W: Wohaish, CM: Chabd ‘l-Mlusi, N: Nijili, SM: South wadi Mlusi, M: Mlusi, NM: North Wadi Mlusi and S: Semhat

Kaolin clay is widely utilized in many facets of ancient and modern societies. Traditional applications are many and some of the more important include ceramics, paper, refractories, paint, plastics, foundry bondants, chemical carriers and many others (Murray, 2000). The present study deals with the assessment of nineteen kaolin clay deposits, of Permian age, located at the Ga'ara Depression, Iraqi Western Desert (Fig.1). Physical minera1ogica1 and chemical properties of each of these deposits are investigated depending on representative bulk samples in order to determine their suitability for a variety of industrial application. Three deposits are densely sampled in order to check their homogeneity viz. South Wadi Mlusi (SM), West Wadi Tayyarah (WT) and South Ghadir 'l Sufi (SS).

GEOLOGICAL SETTING

The Ga'ara Formation is exposed in the Ga'ara Depression only. The Ga'ara Depression is suboval and asymmetrical as the southern rim is much steeper and ragged. It is erosional topographic feature extending in E – W direction (70 × 30 Km) and is located about 50 Km north of Rutba Town, at the Western Desert of Iraq. The area is characterized by fresh and clean surfaces, scarcity of vegetation, abundance of rills, intense drainage and immature soil. These clues indicate that the erosion in the Depression is effective. The area falls within the northern edge of the Arabian Platform. This formation is the oldest exposed unit in the western part of Iraq. Paleobotanical studies revealed that the exposed part (uppermost) of the Ga'ara Formation belongs to the Middle to Upper Permian age (Čtyroky, 1973). Palynological studies (Nader *et al*., 1993 and 1994) showed that the age of the Ga'ara Formation (exposed part and in groundwater well K5-1) is Upper Carboniferous – Lower Permian (Stephanian – Autunian) and Al-Haba *et al*. (1994) accepted Westphalian – Artinskian age. The thickness of the formation in the study area is at least 800 metres but the exposure rarely exceeds 60 meters. The exposed part consists of cyclic repetitions of fluvial sandstone siltstone, silty claystone and claystone (Tamar-Agha, 1986). The sandstone is found as lenses (less than 1 to 25 metres thick; when the channels are stacked on each other) representing ancient channels whereas, the rest form the major lithology representing floodplain deposits (Fig.2). The clays and silty clays of the Ga'ara Formation are one to over 30 metres thick of soft to medium tough, ferruginous in places and multicoloured. The colour is white, grey, yellow, brown, red and violet being uniformly distributed or mottled. Silt and sand admixtures are commonly found increasing especially in the lower part of the clay horizons. Coalfield plant debris and rootlets are also frequent. In few cases the clays are rich with organic content (Sadiq, 1985).

SAMPLING AND METHODS

Nineteen representative bulk samples, about 20 kilograms each, were collected from core-drilled boreholes and trenches to represent the whole of each deposit. All samples were crushed, well mixed and divided into small portions by quartering. Additional samples were collected in order to check the variation of the properties within single deposits. The bulk samples were identified by abbreviations (symbols) for convenience (Fig.1 and Table 1). Below is an outline of the methods used and number of samples.

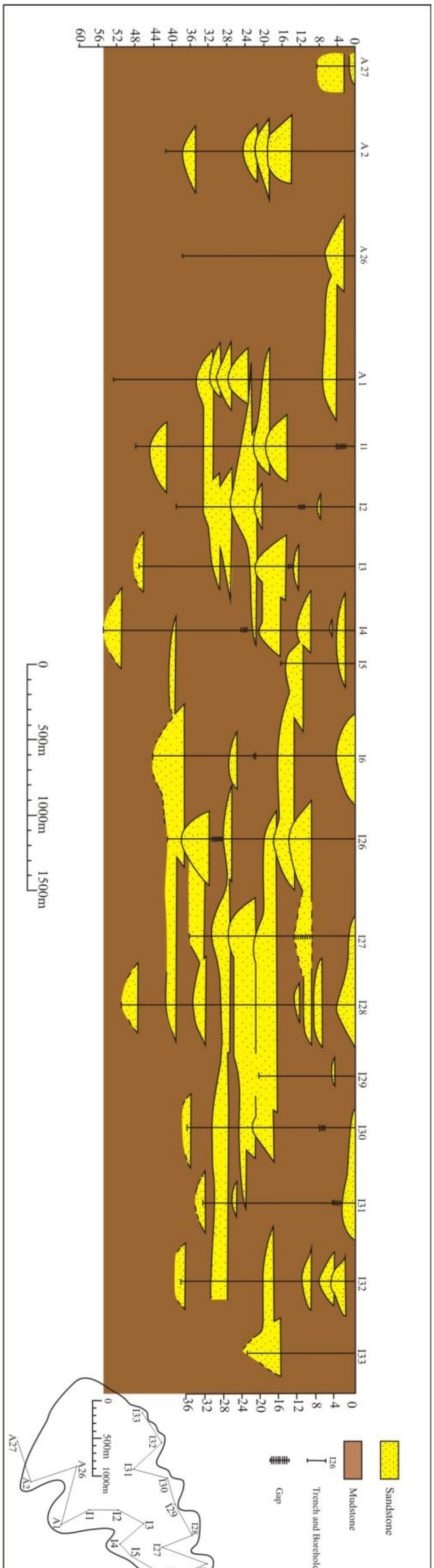

Fig.2: Correlation diagram in the Ga'ara Formation showing the pattern of sedimentary facies distribution

Table 1: Approximate mineral composition (as calculated from the chemical constituents shown in Table 5), organic matter (calculated from L.O.I) and crystallinity index (from X-ray diffractometry, using Hinckley's method, 1963) of the studied Ga'ara deposits

Name of deposit	Symbol	Mineral composition and other constituents				Crystallinity Index
		Kaolinite %	Quartz %	Organic matter %	Others %	
North Wadi Tayyarah	NT	71	21	0.2	7.8	0.4
East Wadi Tayyarah	ET	77	16	0.2	6.8	0.4
East Afaif	EA	82	14	0.2	3.8	0.4
West Wadi Tayyarah	WT	82	14	0.3	3.7	0.4
Tel Afaif	A	76	22	1.0	1.0	0.5
East Duekhla	ED	76	17	0.2	6.8	0.6
Duekhla	D	88	8	0.4	3.6	0.6
Ubairan	U	79	15	0.2	5.8	0.6
Azzlat Al-Agharri	AA	79	14	0.5	6.5	0.4
Wadi Al-Agharri	WA	77	16	0.4	6.6	0.5
South Ghadir 'l Sufi	SS	61	34	0.2	4.8	0.3
Ghadir 'l Sufi	SU	64	27	0.3	8.7	0.4
Wohaish	W	56	37	0.4	6.6	0.4
Chabd 'l-Mlusi	CM	59	34	0.4	6.6	0.4
Nijili	N	63	28	0.4	8.6	0.5
South wadi Mlusi	SM	71	21	0.2	7.8	0.5
Mlusi	M	52	41	0.2	6.8	0.3
North Wadi Mlusi	NM	63	34	0.3	2.7	0.4
Semhat	S	78	20	0.1	1.9	0.3

- **Mineralogy**

All collected samples were studied by X-ray diffraction (XRD) at the laboratories of the Geological Survey of Iraq, seven samples by transmission electron microscopy (TEM) at the laboratories of the University of Hull – United Kingdom, three samples by scanning electron microscopy (SEM) at the Habibiya Laboratories of the Oil Exploration Company (OEC) and ten samples by differential thermal analysis (DTA) at the Iraqi Scientific Research Council. For X-ray diffractometry, the fraction finer than 53 µm of all samples is studied using unoriented mounts whereas the fraction finer than two microns is studied using both oriented and unoriented mounts. Clay minerals are best analyzed on oriented mounts whereas unoriented mounts are prepared in order to determine the crystallinity index using XRD patterns of the < 2 µm size fraction of unoriented clay samples (Hinckley, 1963). Some of the mounts are glycolated and heated (350° and 550 °C) in order to check the clay mineralogy. The mounts were run in Phillips X-ray diffractometer PW 1965/60 (20 mA, 4.0 kV, Cu-Ni Kα, at 1° centimetre/minute, 1° 2θ-/minute speed). DTA is made by heating dried finely ground extracted samples and extracted clay fraction < 2 µm up till 1000 °C using a constant rate of 8 °C/minute.

- **Physical Properties**

– **Particle size distribution:** The distribution of particle size was carried out for all bulk samples using hydrometer method as specified in the ASTM standard specification D7928-17 (2017). In addition to the nineteen bulk samples, 167 other samples are selected from three deposits namely West Wadi Tayyarah (28 samples), South Ghadir '1 Sufi (26 samples) and South Wadi Mlusi (113 samples) in order to check the homogeneity of the three deposits in respect to their particle size distribution.

– **Colour measurements:** All samples are oven-dried overnight at temperature 105 ºC, ground and pelletized in order to check their whiteness and brightness. For measurement of whiteness the pellets are compared with a magnesium oxide standard (99.5% whiteness) at wavelength 6860 Å using Dr. Lange Colorimeter. GE Brightness was carried out on the samples after pelletization by using Ernst Leitz Reflection Meter at wavelength 458 nm. Colour measurements are carried out on 14 selected samples, by obtaining a measurement of the amount of light reflected from the surface of the powdered sample relative to a standard (magnesium carbonate). The E.E.L. Reflectance Spectrophotometer was used with nine filters from red to violet and the results are displayed on a graph to obtain spectrophotometric curve system. Colour measurements were made for the unfired samples after separation of the fraction coarser than 53 µm and for the same samples after firing (1000 °C).The samples were ground and pelletized using a specially made powder press (based on the Zeiss Elerpho Press) without the use of a binder.

– **Plasticity index** (Atterberg's limits)**:** This is determined using the standard method as specified in the ASTM standard specification D424 (1971).

– **Nature of slaking:** It was carried out on lumps of few centimetres in diameter. The lumps were placed in a beaker filled with water and the time required for disintegration is recorded.

- **Chemical Analysis**

All samples were analyzed in the laboratories of the Iraq Geological Survey (GEOSURV-Baghdad) for the following constituents: SiO_2, Al_2O_3, Fe_2O_3, TiO_2, Na_2O, K_2O, CaO, MgO, MnO and SO_3 (mostly by atomic absorption spectrophotometry-AAS). Loss on ignition (L.O.I.) is determined on two stages; by heating to 350 ºC and 1000 °C. Determination of pH of the aqueous suspension of the samples were carried on 10% (by weight) suspension, which were allowed to stand for one hour after mixing the powdered samples with distilled water.

PROPERTIES OF THE GA'ARA KAOLIN CLAYS

The evaluation of a kaolin clays deposits for industrial applications depends largely on their properties to single out the type of processing necessary to produce the required product or may even help in choosing the appropriate technology for refining or manufacturing. Some of these properties are important for many applications but others are only appropriate for single application.

- **Mineralogy**

X-ray diffractometry reveals that the major clay mineral present in the Ga'ara clays is kaolinite with occasional traces of illlte (Fig.3). The main non-clay minerals are quartz, iron oxihydroxides and anatase. Quartz is invariably present in all size fractions, (sand, silt and clay). Iron oxyhydroxides are mostly goethite with minor amount of hematite. Their presence gives the yellow, brown, red and purple colours to the clays. Organic matter, in the form of plant remains, is evident from field observation and gives grey tint to most of the samples.

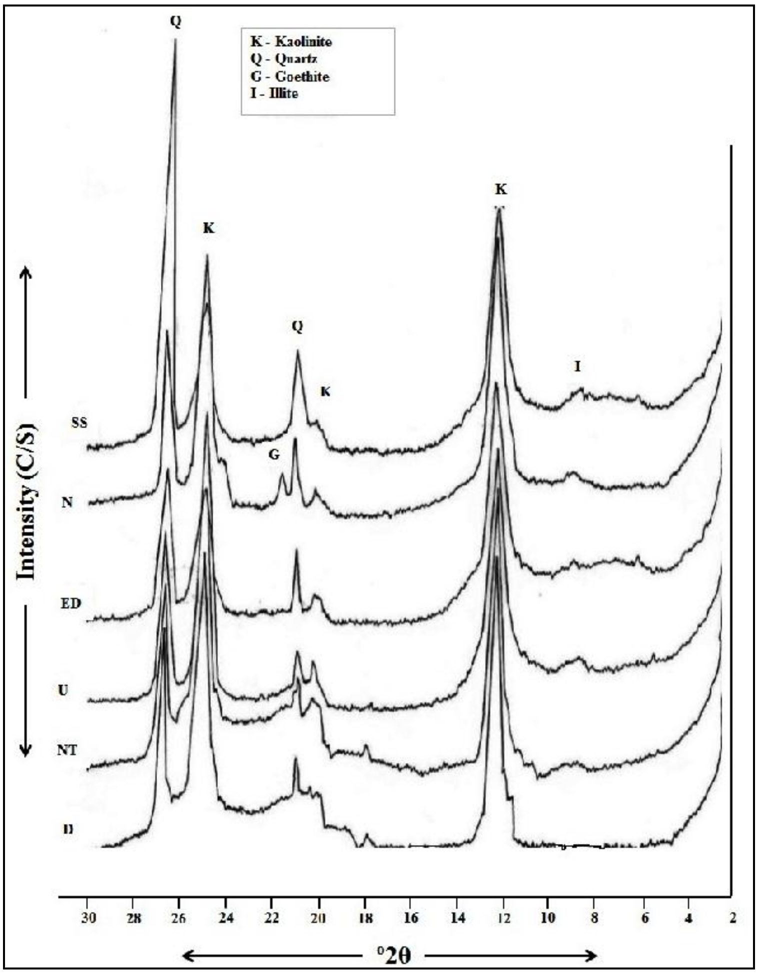

Fig.3: XRD pattern of the unoriented clay fraction (< 2 µm) of some representative samples of the Ga'ara kaolin clays

The crystallinity index of the Ga'ara kaolinite ranges from 0.3 to 0.8 which represents poor degree of crystallinity. On this scale, a degree of crystallinity of zero represents kaolinite with undeveloped crystal faces, whereas a degree of crystallinity of approximately two represents kaolinite with nearly perfectly developed crystal faces. Both SEM and TEM studies support the XRD reau1ts concerning the poor development of kaolinite crystal faces (Figs.4 and 5). The values of crystallinity index of the Ga'ara kaolinites correspond with the relatively high cation exchange capacity (13 to 19 meq/100 gm). The nature of the diffractograms of the investigated clays compares with an intermediate state of disordering. Tamar-Agha (1991) found that the degree of disorder in the kaolinite of the Ga'ara clays varies considerably within a single clay horizon depending on its position within the depositional cycle, i.e. clays of the upper part of the horizon show higher degree of disorder.

The DTA curves are basically similar, with little variation can be noticed in the size of the peaks and the temperature at which they occur. The main peaks are:

− **Peak (A):** A small endothermic peak occurs at temperature range 70 to 107 °C which results from the removal of the non-constitutional water and is related to the water sorbed onto the surface of the fine clay particles. The variation in the temperature of loss may be attributed to the presence of broken bond water which may not be lost until a sufficiently high temperature is reached.

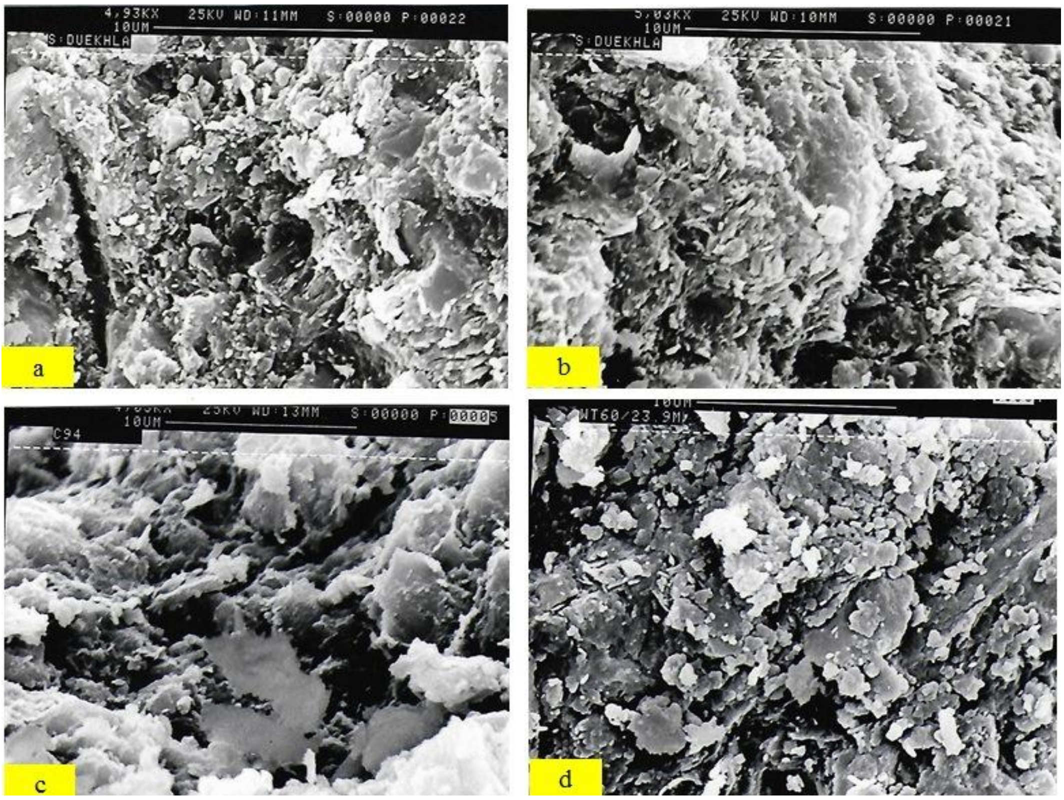

Fig.4: SEM photographs of the Ga'ara kaolin clays from Duekhla deposit (a and b) and West Tayyarah deposit (c and d)

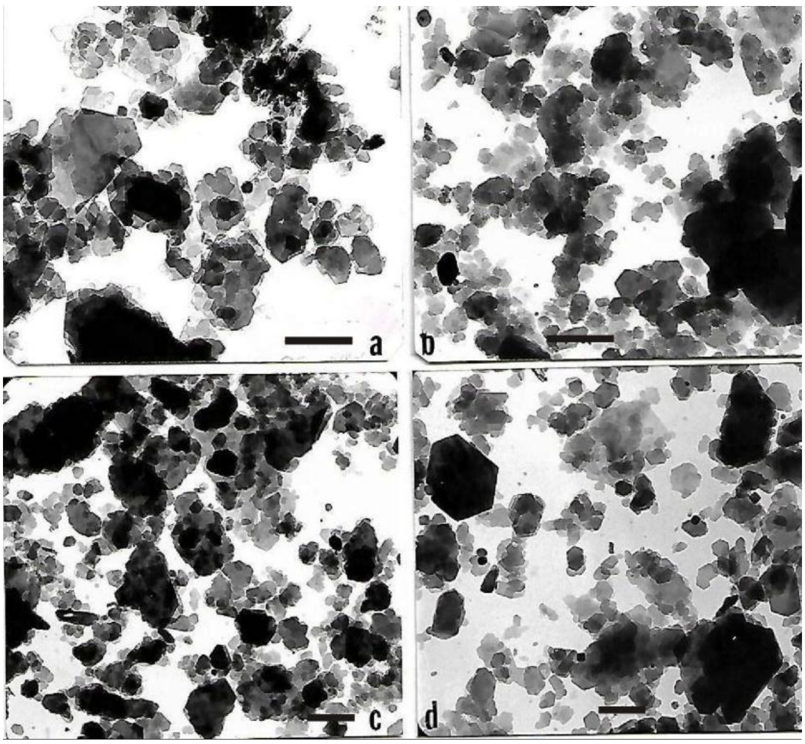

Fig.5: TEM photographs of some of the investigated clays showing stacked plates of poorly-developed pseudohexagonal kaolinite. **a)** Duekhla, **b)** Semhat, **c)** Azzlat Al-Agharri and **d)** Duekhla. Bar scale = 1 µm

– **Peak (B):** Another small endothermic peak occurs at temperatures between 320° and 330 °C. This peak is attributed to the loss of structural water from a small amount of goethite present in the samples.

– **Peak (C):** A major endothermic peak occurs at temperatures between 560 °C and 575 °C. It is the main characteristic endothermic peak for kaolinite which corresponds to the expulsion of the bulk of water from its lattice.

– **Peak (D):** This exothermic peak lies between 940 °C and 950 °C. The slight difference might be due to the presence of impurities such as iron oxide and alkalis which act as fluxes and thus reduce the temperature of melting.

The variation in the magnitude of the DTA peaks could be attributed to differences in particle size which is expected from clays collected from different deposits. Since the amount of carbonaceous material in the clays is very low, no pronounced peaks are recognized for the combustion of this material. The only peak recognized appeared at a temperature of 280 °C and even this is small (Fig.6). Generally all coloured clays show little variation in their firing properties.

The approximate mineral composition is calculated using two methods. The first is from the chemical analyses. The theoretical chemical formula of kaolinite is 39.5 % Al_2O_3, 46.5% SiO_2 and 14.0% H_2O. The alumina content is used as a basis to calculate the kaolinite percentage and the rest of the silica as quartz. The second procedure for determining the kaolinite percentage is taken from loss of ignition. Kaolinite loses its combined water at temperatures well below 1000 ºC. From weight reduction (resulting from the loss of water on heating) it is possible to determine the percentage of kaolinite present. The procedure is as follows: samples were dried overnight in an oven at 105 ºC to remove all absorbed water. Approximately 2 g of each sample was accurately measured (to four decimal places) into a previously ignited silica crucible and heated to 350 ºC in a furnace for one hour. The crucible and contents were then cooled in a desiccator and again accurately weighed. The loss in weight is attributed to combustion of organic matter. The sample and the crucible were then returned to the furnace and heated to 1000 ºC for at least one hour. After the furnace had cooled the crucible and content were removed, allowed to cool in a desiccator and once again accurately weighed. The loss in weight on ignition between 350 ºC and 1000 ºC is taken as an indication of the amount of kaolinite in the sample. Both weight loss values are expressed as percentage of dried sample.

For the investigated samples, where kaolinite is the predominant clay mineral, the test is useful in obtaining a general idea about the kaolinite content and the organic matter present. The kaolinite content of the studied kaolin clay deposits ranges from 88% in Duekhla deposit to 52% in Mlusi deposit (Table 1). Using the same formula above, the free silica left is calculated as quartz, despite that this formula represents well-ordered kaolinite and the studied samples show some disorder in their crystal lattice. The calculated quartz content in the studied clays ranged from 41% in Mlusi deposit to 8% in Duekhla deposit. Other minerals present are minor being mostly iron oxihydroxides and anatase except in the Nijili clay deposit whereby they form about 8% of the sample. This deposit represents coloured kaolin clay and hence has relatively high content of iron oxides.

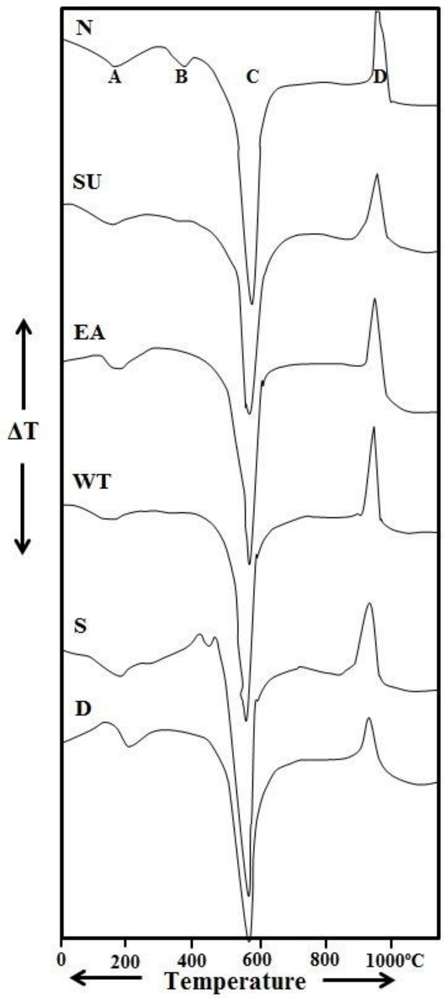

Fig.6: DTA curves for some representative kaolin clay samples

- **Physical properties**

– **Particle size distribution:** A measure of particle size distribution of the raw material and the product is important for almost all applications of kaolin. It governs its use in paper (filler and coating grade), ceramic (partially controls plasticity), paint (affects technological properties), rubber and plastic industries (different reinforcing properties). The results show that there is a large variation in the grain size of the samples (Table 2). Detailed investigations on the particle size distribution of three deposits; namely South Wadi Mlusi (SM), West Wadi Tayyarah (WT) and South Ghadir '1-Sufi (SS), showed that there is a large variation, even within the deposit itself, indicating the sediments heterogeneity (Fig.7, Table 3).

On the basis of particle size, the investigated kaolin clay deposits can be classified into four groups: very fine, fine, medium and coarse clays. In the very fine grained clays, the percentage of the particles coarser than 20 µm does not exceed 5% of the total sample. This group includes eight deposit samples: ET, EA, WT, ED, D, U and S. The percentage of the < 2 µm fraction in clays of this group ranges from 82.9% in sample D down to 77.5% in sample S and the mean of particles finer than 5 µm is 90% (Fig.7). This group of clays has exceptionally high percentage of fine materials and small amount of coarse fraction which means that little or no processing of the clay would be necessary to obtain pure kaolin and increases the industrial value of the material considerably.

Table 2: Size class (using the present classification) of each deposit studied of the Ga'ara kaolin clay and the percentages of materials in each class interval

Name of deposit	Symbol	Size class	>20 µm	10 – 20 µm	5 – 10 µm	2 – 5 µm	-2 µm	-1 µm
North Wadi Tayyarah	**NT**	Very fine	4.9	3.9	4.1	9.6	77.5	69.3
East Wadi Tayyarah	**ET**	Very fine	2.3	2.0	6.0	8.0	81.7	77.2
East Afaif	**EA**	Very fine	2.1	2.1	5.0	8.3	82.5	78.5
West Wadi Tayyarah	**WT**	Very fine	4.9	3.6	3.9	9.4	78.2	70.4
Tel Afaif	**A**	Medium	12.7	5.1	11.0	14.1	57.1	45.3
East Duekhla	**ED**	Very fine	2.1	2.1	4.1	10.5	81.2	67.2
Duekhla	**D**	Very fine	4.1	2.8	2.9	7.3	82.9	68.9
Ubairan	**U**	Very fine	3.9	2.9	3.1	7.7	82.4	65.3
Azzlat Al-Agharri	**AA**	Medium	11.3	12.1	13.6	13.9	49.1	37.2
Wadi Al-Agharri	**WA**	Medium	12.3	8.4	8.2	14.8	56.3	49.2
South Ghadir ‘l Sufi	**SS**	Fine	5.1	3.2	6.9	17.8	67.0	58.1
Ghadir ‘l Sufi	**SU**	Fine	5.8	4.5	7.2	16.2	66.3	60.0
Wohaish	**W**	Coarse	17.0	11.5	11.8	12.9	46.8	35.2
Chabd ‘l-Mlusi	**CM**	Coarse	18.2	13.0	11.8	11.1	45.9	33.3
Nijili	**N**	Medium	10.9	8.5	11.3	15.2	54.1	40.7
South Wadi Mlusi	**SM**	Medium	11.3	11.6	9.0	16.4	51.7	31.7
Mlusi	**M**	Medium	14.0	8.9	10.5	15.6	51.0	34.1
North Wadi Mlusi	**NM**	Fine	5.5	4.2	6.6	8.5	75.2	61.7
Semhat	**S**	Very fine	0.5	5.5	9.5	18.6	65.9	61.1

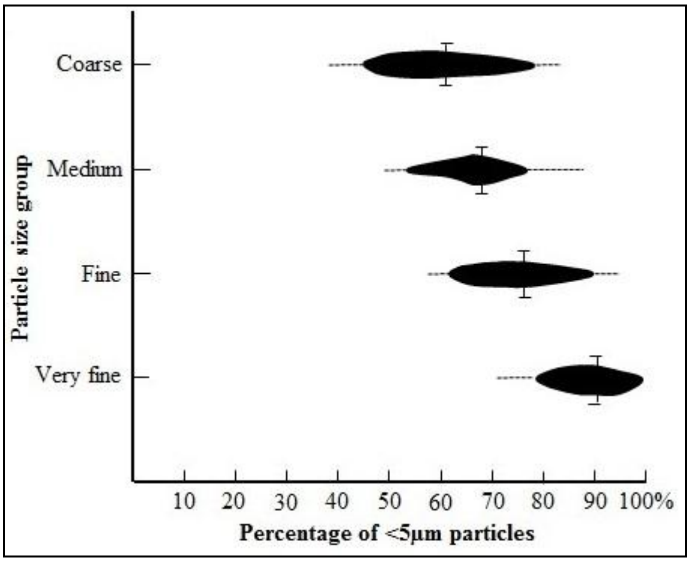

Fig.7: Distribution of particles finer than 5 microns in the different particle size groups

Table 3: Summary of the particle size distribution of three kaolin clay deposits SM, WT and SS

Name of deposit	Classification					>20 µm			10 – 20 µm			5 – 10 µm			2 – 5 µm			<2 µm			< 1µm			Sorting*
	Class of the deposit	Number of samples in each class																						
		Very fine	Fine	Medium	Coarse	Mean	Maximum	Minimum	Mean	Maximum	Minimum	Mean	Maximum	Minimum	Mean	Maximum	Minimum	Mean	Maximum	Minimum	Mean	Maximum	Minimum	
South Wadi Mlusi (SM)	Medium	16	29	28	40	11.3	50	2	11.6	16.5	1	9	23	3	16.4	40	4	51.7	53.5	2	31.7	68	9	Poorly sorted
West Wadi Tayyarah (WT)	Very fine	17	6	2	3	4.9	19.1	0.1	3.6	20.0	0	3.9	10	0	9.4	23.3	1.1	78.2	97.8	54.2	70.4	91.9	46.1	Poorly sorted
South Ghadir 'l Sufi (SS)	Fine	18	7	-	1	5.1	15.1	0.06	3.2	13.2	0	6.9	12.1	1.0	17.8	25.1	9.3	67.0	84.9	51.1	57.1	73.5	45.6	Medium sorted

In fine grained clays, the percentage of the particles coarser than 20 µm ranges from 5 to 10% of the total sample. This group includes three samples SS, SU and NM. The percentage of the < 2µm in size decreases as the percentage of coarse material increases (Table 2) ranging from 75.2% in sample NM to 66.3% in sample SU. The materials finer than 5 µm are considerably lower than those in the previous group but yet, their mean values are still high (76%). In medium grained clays, the percentage of the particles coarser than 20 µm ranges from 10-15% of the total sample. Samples A, AA, WA, N, SM and M belong to this group, and they contain particles finer than 2 µm from 57.1 % in sample A to 49.1% in sample AA (mean of particles finer than 5 µm is 67.5%). In coarse grained clays, the percentage of the particles > 20 µm is more than 15% of the total sample. Samples W and CM belong to this group which contains the lowest percentage of material in the 2 µm size fraction (60%).

For ceramic purpose Budnikov (1964) classified clays according to the content of particles less than one micron size into highly dispersed (60%), dispersed (20 – 60 %) and coarsely dispersed (20%). Accordingly, nine of the Ga'ara clays are highly dispersed (NT, ET, WT, ED, D, U, NM and S) and ten are dispersed (A, AA, UA, SS, SU, U, CM, N, SM, and M).

– **Colour:** Another important factor used in assessing kaolin deposits is their colour. The original colour of the clay deposit is important to such industries as paper, rubber and paint whereas the colour after firing is important to other industries such as ceramics. The variation in the colour of the clays and their brightness is a function of several variables but the major factors are iron oxihydroxide and organic matter contents. Titania and particle size distribution also affects the colour parameters but to a lesser degree (Murray, 2007).

The majority of iron oxihydroxide is present as coating around clay particles or disseminated impurities and small amount may be found within the structure of kaolinite (Tamar-Agha, 1986; Al-Sayegh *et al*., 1994; Jassim *et al*., 2009). Spectrophotometeric graphs (Figs.8 and 9) show noticeable drop in reflectivity towards the short wavelength end of the spectrum for clay samples which contain higher levels of iron oxide. Iron oxihydroxide occurs as goethite and hematite which are considered as heavy absorbers of light in the violet-green region of the spectrum and absorption decreases towards the red end (Mustafa, 1981). This is illustrated by the steepness of the spectrophotometric curves in samples with higher iron oxides.

Organic matter is present as plant remains and its content in general vary greatly. Even a little content of organic matter induces a grey colour to the clay. It is expected that organic matter absorb light to a greater extent than kaolinite, thus bringing about a decrease in reflectivity as its content increases. Other minerals present, particularly quartz which is present in all samples and sometimes gypsum (in some near-surface samples), are expected to have some effect in decreasing the reflectivity of the clays.

The Ga'ara kaolin clays were divided on the basis of iron oxide content into white and coloured kaolin clays (Zurek and Knapp, 1960). The maximum content of iron oxide in the white clays was taken as 2%. Nevertheless though they are referred to as white clays, they are frequently grey or pink. According to this grouping, the studied samples are white except samples N and W. Whiteness of the bulk samples ranges from 64.1% in N to 85.2% in EA (Table 4). Four samples can be classed as white clays (whiteness 80% or more) which are ET, EA, SM and S; 13 samples can be classed as off-white clays (whiteness 70% to 80%) which are NT, WT, A, ED, D, U, AA, WA, SS, SU, CM, M, NM; and only two samples can be classed as coloured clays (whiteness less than 70%) namely W and N. Brightness of these samples ranged from 52% in N to 71.7% in EA (Table 4).

From the spectrophotometeric graphs it is also possible to divide the studied clay samples into three groups namely white, near-white and coloured (Fig.8). The colour of a single deposit varies considerably laterally as well as vertically, that is they are heterogeneous even within one deposit. The white clays have reflectivity ranging from 62 to 75% at the violet end and 70 to 90 % at the red end of the spectrum (Fig.8a). The reflectivity of the near-white clays ranges from 50 to 60 % at the violet end and 71 to 75 % at the red end of the spectrum (Fig.8b). The coloured clays show reflectivity below 60% in the violet region and often less than 70% in the red region and often less than 70% in the red region (Fig.8c).

In ceramics, the fired-clay colour is more important than the colour of the unfired clay. For the manufacture of pottery and white ware, white fired clays are often required. The colour of fired clay depends mainly on the proportion of iron oxide it contains, but firing temperature, particle size, degree of vitrification and other factors can also modify the colour (Ryan, 1978 and Al-Kaiysi, 1989). The presence of CaO and MgO may whiten the clay. The majority of the white Ga'ara clays show an improvement in colour on firing (Fig.9) whereas those containing relatively higher iron oxide show a reduction in overall reflectance. Calcination of clays containing organic matter impurities improves its colour parameters but undoubtedly affects other properties too.

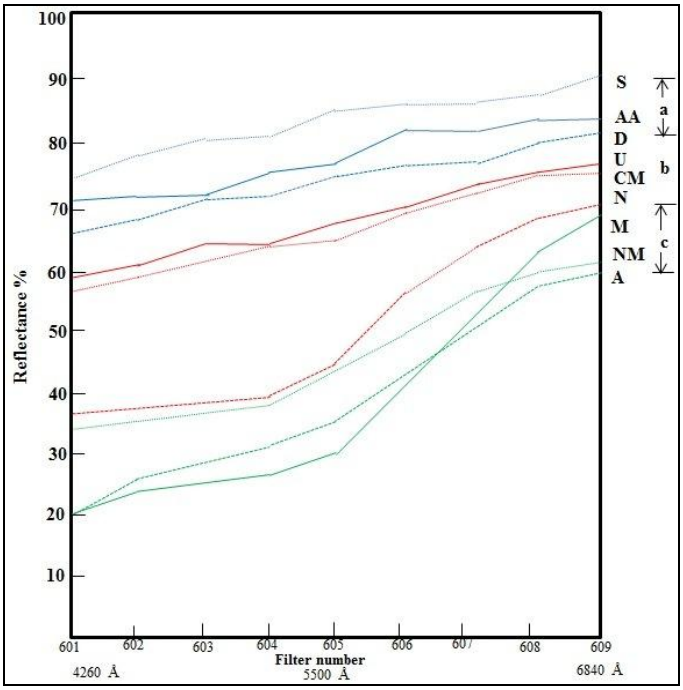

Fig.8: Spectrophotometric curves of the fine fraction (-53 µm) for some of the unfired samples; (a) white clays, (b) near-white and (c) off-white

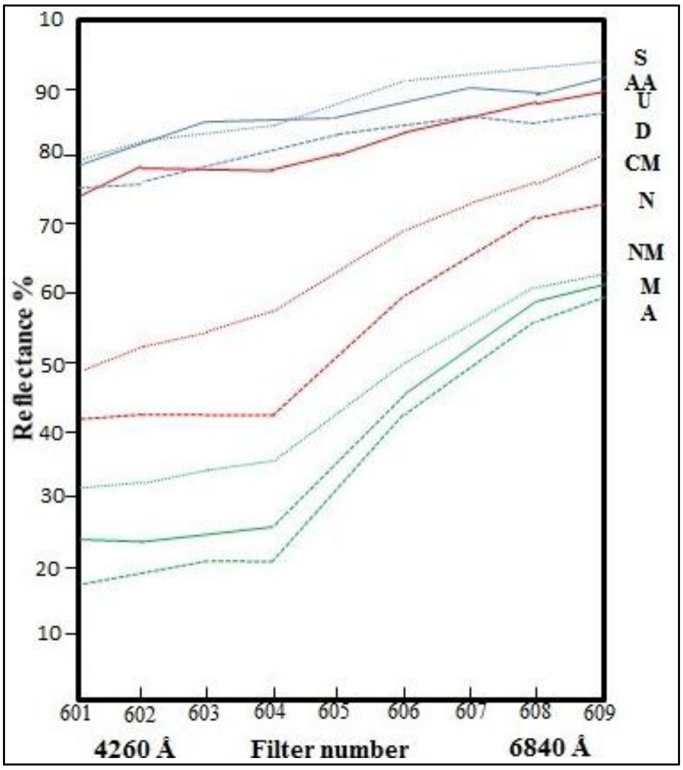

Fig.9: Spectrophotometric curves of the fine fraction for some of the fired samples

Table 4: Colour and plasticity measurements of the studied samples of the Ga'ara kaolin clay deposits

Symbol of deposit	Colour Measurements		Plasticity		
	Whiteness	Brightness	Liquid Limit	Plastic Limit	Plasticity Index
NT	74.8	54.8	50.8	25.8	25.0
ET	84.8	71.3	52.2	26.3	25.9
EA	85.2	71.7	53.6	26.9	26.7
WT	75.1	56.4	53.7	25.5	28.2
A	72.0	53.0	50.4	25.3	25.1
ED	73.3	54.5	54.3	25.2	29.1
D	72.7	54.1	57.3	27.1	30.2
U	73.2	55.2	55.8	26.0	29.8
AA	78.2	54.3	48.8	25.6	23.2
WA	79.8	68.0	50.9	26.5	24.4
SS	71.2	53.8	42.1	23.8	18.3
SU	72.6	54.0	49.1	26.1	23.0
W	68.8	53.0	44.2	24.1	20.1
CM	72.3	54.5	47.9	25.5	22.4
N	64.1	52.0	48.2	25.3	22.9
SM	80.4	63.0	47.8	24.5	23.3
M	73.2	55.0	45.3	24.0	21.3
NM	74.7	70.1	45.6	23.5	22.1
S	80.1	68.2	50.6	23.4	27.2

– **Plasticity:** The plasticity of clays gives an indication of their suitability for commercial exploitation in the ceramic industries. Budnikov (1964) classified clays on the basis of plasticity index, for ceramic industries, into five groups. The plasticity index of the investigated clays ranged from 21.3 to 30.8 (Table 4) that is plastic to superplastic (according to Budnikov, 1964 classification, plasticity index of plastic clay is 15 to 25 whereas that of superplastic clay is more than 25). The results obtained in this study vary from one sample to another because there are many factors which affect the plasticity of the clay such as particle size distribution, non-clay mineral constituents and the presence of organic matter. Plasticity index is directly proportional to the kaolinite content (Fig.10a). The presence of non-plastic accessory minerals such as quartz, not surprisingly has a considerable effect on the clay (Fig.10b). Mineralogical investigations revealed that quartz is invariably present in the Ga'ara kaolin clays in all size grades though in different proportions (Al-Youzbaky, 1989). Plastic index and plastic limit values obtained can be correlated with the percent of clay present as indicated by the values of loss on ignition. Superplastic clays are very fine and have kaolinite content more than 75%. The majority of the Ga'ara kaolin deposits resemble the ball clay of the United Kingdom. They are plastic to superplastic, fine to very fine, grey in colour and fires to off-white to buff.

– **Nature of slaking:** This test indicates the binding strength of the clay particles and the degree of consolidation of the clay mass. It is noticed that all samples disintegrate in water shortly after their immersion; that is within two to four minutes. The nature of slaking of the Ga'ara kaolin clays can therefore be referred to as quick.

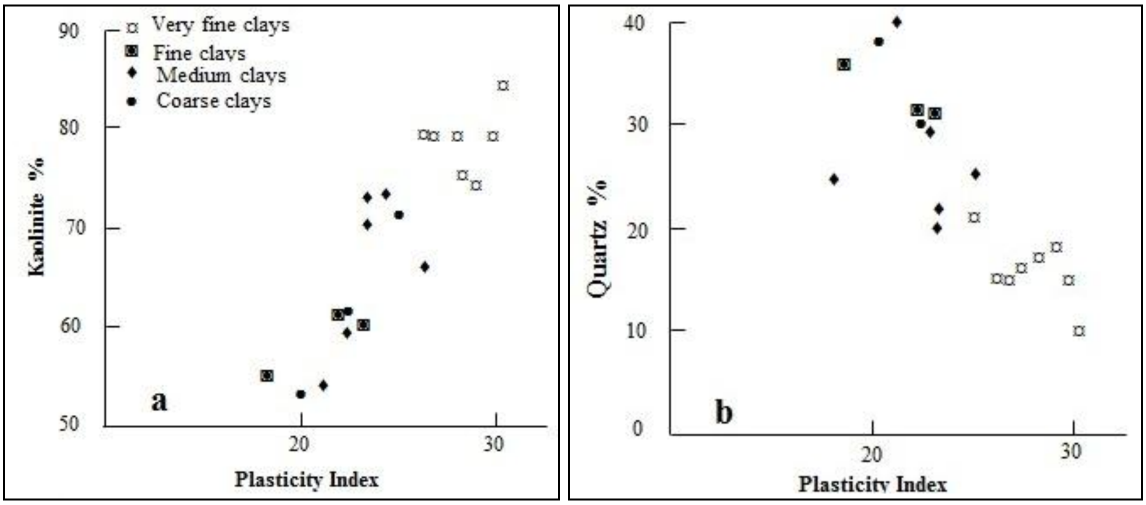

Fig.10: Covariation of kaolinite percentage (**a**) and quartz percentage (**b**) with plasticity index in the size classes of the Ga'ara kaolin clay deposits

- **Chemical properties**

– **Chemical constituents:** The chemical composition of a clay deposit is important for certain application as it becomes the chief factor which governs its suitability in certain industries such as ceramics and refractories. The main oxides of the studied clays are SiO_2 and Al_2O_3 (Table 5). Significant amounts of Fe_2O_3 are present in some samples ranging from 1.0 to 6.5%. The majority of the iron oxide is present as coating around the clay particles and/or as separate mineral grains (as goethite, hematite and amorphous Fe-hydroxides and oxihydroxide) and not in the structural lattice of the clay minerals (Al-Youzbaky, 1989; and Al-Sayegh *et al*., 1994). The total amount of TiO_2 is approximately constant and range between 0.9% and 1.92. These values of titania are typical of sedimentary kaolin deposits (Murray, 2007). The total alkalis (Na_2O, K_2O, CaO and MgO) which behave as fluxes during firing, are generally low (around 1.5%). MnO content is rather insignificant ranging from 0.01 to 0.03%. Sulphates (SO_3) are low in fresh samples and fairly high in samples close to the surface where gypsum increases considerably

The Ga'ara kaolin clays studied are heterogeneous consisting of clays of different proportions of chemical constituents. Tamar-Agha and co-workers (1991) showed that each of these deposits contains variable amount of admixture which affects its resultant chemical constituents. For instance in West Wadi Tayyarah deposit the alumina content ranges in the reserve from 20.0 to 38.7 wt.% Al_2O_3. The alumina content in the studied clays ranges from 20.5 to 34.7% (Table 5). On the basis of alumina content, the deposits can be classified into fat clays (> 30% Al_2O_3) such as ET, EA, WT, A, ED, D, U, WA and S, moderately fat clays (25 to 30% Al_2O_3) such as NT, AA, SU, CM, SM, and NM and lean clays (20 to 25% Al_2O_3) such as SS, W, N and M. Each of these clay deposits contains both fat and lean clay horizons.

Some of the chemical constituents of the studied clays are not favorable in some industries; e.g. iron oxides in refractories and whiteware ceramics, and free silica in paper manufacturing and some pharmaceuticals. These constituents can be reduced by two main methods: the elimination of beds with higher content of such deleterious constituents during mining operations or post-mining mineral dressing and upgrading such as washing or high intensity electromagnetic separation.

Table 5: Chemical constituents (wt.%), pH value of the suspension and cation exchange capacity (c.e.c meq./100 g) of the analyzed samples of the Ga'ara kaolin clay

Deposit Symbol	Chemical Constituents												pH	c.e.c
	Al_2O_3	Fe_2O_3	SiO_2	TiO_2	LOI. 350 °C	LOI. 1000 °C	SO_3	Na_2O	K_2O	CaO	MgO	MnO		
NT	27.9	1.8	53.1	1.2	0.2	10.20	0.1	0.7	0.2	0.9	0.5	0.01	8.2	16
ET	30.3	1.1	51.3	1.5	0.2	11.15	<0.07	0.4	0.1	0.2	0.2	0.01	8.2	17
EA	31.2	1.2	51.0	1.7	0.2	11.20	0.09	0.4	0.1	0.5	0.2	0.02	8.3	16
WT	32.2	1.8	51.8	1.3	0.3	10.80	<0.07	0.5	0.3	0.2	0.3	0.03	8.2	17
A	30.1	1.3	55.2	0.9	1.0	10.30	<0.07	0.4	0.3	0.4	0.3	0.02	8.4	12
ED	30.1	1.8	52.1	1.2	0.2	10.50	<0.07	0.5	0.3	0.3	0.3	0.02	8.2	17
D	34.7	1.1	49.0	1.3	0.4	12.10	<0.07	0.4	0.3	0.4	0.3	0.02	8.2	17
U	31.2	1.4	51.2	1.4	0.2	11.20	0.09	0.3	0.4	0.3	0.3	0.02	8.1	16
AA	28.9	1.0	54.9	1.9	0.5	10.30	0.09	0.6	0.3	0.3	0.2	0.02	8.3	13
WA	30.4	1.8	51.5	1.3	0.4	10.60	0.10	0.5	0.4	0.4	0.3	0.02	8.1	13
SS	24.0	2.2	61.8	1.2	0.2	7.90	<0.07	0.4	0.5	0.3	0.3	0.03	8.3	14
SU	25.1	2.1	59.3	1.1	0.3	8.70	<0.07	0.4	1.1	0.5	0.6	0.02	8.2	15
W	22.0	1.9	63.1	1.1	0.4	7.80	<0.07	0.4	0.6	0.3	0.4	0.03	8.4	12
CM	25.3	1.5	58.2	1.4	0.4	8.90	0.09	0.4	0.6	0.2	0.4	0.01	8.3	10
N	24.8	6.5	57.1	1.2	0.4	8.80	<0.07	0.4	0.3	0.2	0.5	0.02	8.4	14
SM	28.2	1.4	54.1	1.4	0.2	10.70	0.1	0.6	0.1	0.3	0.4	0.02	8.1	13
M	20.5	2.5	65.1	1.0	0.2	7.80	<0.07	0.4	0.2	0.4	0.4	0.03	8.3	14
NM	25.1	2.0	59.5	1.2	0.3	8.90	<0.07	0.4	0.6	0.3	0.3	0.02	8.2	16
S	31.1	1.3	53.0	1.2	0.2	11.20	0.09	0.3	0.7	0.2	0.4	0.03	9.1	17

– **pH of suspension:** An aqueous suspension of a kaolin clay is usually slightly alkaline. It is important for rheology of flow behavior of aqueous suspension (e.g. in slip casting and paper industry (Murray, 2007). The pH of the studied samples ranged from 8.1 to 8.4 except one sample taken from Semhat deposit whereby the reading was 9.1 (Table 5).

– **Cation exchange capacity:** The cation exchange capacity (CEC) of the studied samples ranges from 13 to 19 m.eq./100 g (Table 5). This range of CEC values is rather moderate compared to the ball clays and fireclays ranging from 15 to 40 m.eq./100 g (Ryan, 1978).

POTENTIALS FOR INDUSTRIAL UTILIZATION

Kaolin is used in wide range of industries such as paper, paints, rubber, plastics, ceramics, refractories, cement, pesticides, textiles, adhesive, pharmaceuticals, catalysts, aluminium (and aluminium compounds) and many others. The majority of the investigated clay samples from the Ga'ara area have the potential to be used in different consuming industries but the amount of beneficiation required will vary. Recommendations regarding the possible industrial utilization of the studied clay deposits will be based on the assumption that beneficiation is to be kept to a minimum to avoid practical complications and additional expenses. For this reason only, clays of the Ga'ara Depression, whose properties bring it close to a particular specification, are recommended for specific end-use. Kaolin clay of the Ga'ara Formation is exploited at the present time only from the Duekhla quarry in the Ga'ara Depression. Exploitation started since 1981 and it is mainly consumed, in the manufacturing of white cement (for over two decades and stopped now), ceramics, refractories and paper (as filler).

- **Paper manufacturing**

In this industry kaolin clay is used both as filler and for coating. The most important characteristics of the clay in paper manufacturing are particle size, brightness (off-white or near-white kaolin), particle shape, abrasiveness and some rheological properties (Murray, 2007). Kaolinite is the most widely used mineral in paper manufacturing as filler and coating. It improves paper gloss, smoothness, brightness and opacity, and it improves printability (Bundy and Ishley, 1991). Two types of kaolin clay can be used in this industry: hydrous and calcined (Hartman, 1987).

Milled Duekhla kaolin is currently used in paper industry although there are some reservations to its application in such industry for its low brightness and high abrasiveness. Grain diminution to below 44 microns alone (the procedure currently used in the treatment of Duekhla Kaolin) is unsatisfactory because undesirable materials (quartz, anatase and organic matter) remain in the product and impair its brightness and abrasiveness. It is therefore believed that upgrading of Duekhla kaolin clay by water-washing would improve largely on its properties.

The studied Ga'ara clay deposits show that the kaolinite is poorly crystallized with moderate range of cation exchange capacity and thus have more suitable properties for paper industry. The investigations carried out in the present study show that eight of the clay deposits have potentiality to be used in paper industry namely: East Wadi Tayyarah (ET), West Wadi Tayyarah (WT), East Ataif (EA), East Duekhla (ED), Duekhla (D) Ubairan (U), South Wadi Mlusi (SM) and Azzlat 'l-Agharri (AA). It is unlikely that such deposits are suitable in paper manufacturing without upgrading. Hence detailed studies on upgrading to suite such industry are needed.

Improvement in colour properties and abrasiveness, following removal of the coarse size, is noticed during washing of the samples and separating the clay fraction (-2 µm) for XRD analysis. The colour of the smears prepared from the fines (-2 µm) is noticeably whiter and brighter than smears prepared from the bulk samples. The XRD results supported this as the anatase peaks are absent and quartz peaks were largely reduced. The deleterious materials, such as quartz, anatase and organic matter, seem to concentrate in the coarse fraction. Calcination or oxidation of clays can also be considered in order to remove the organic matter.

- **Ceramic products**

Specifications for clay used in the manufacture of ceramics are not generally as precise as those relating to clays used in the more sophisticated industries. The investigated samples possess suitable plastic properties (Fig.l1). They show a wide range of reflectivity of fired samples due to variations in chemical composition. Most of these clays can be considered as relatively good quality ceramic raw materials and are suitable for a variety of applications within this diverse industry.

The suitability of the Ga'ara kaolin clay, especially the Duekhla deposit, for ceramic industry is much more tackled in comparison to their use in any other industries (Zurek and Knapp, 1960; Mustafa, 1981; Jindy and Ibrahim, 1982; Al-Azzawi and Aly, 1983; Al-Kaiysi, 1989; Al-Mashaykhi, 1991; Tamar-Agha, 1997; Nasir, 2008). These studies dealt with the assessment of these clays for earthenware; stoneware and porcelain (including electro-porcelain and bone china) by using the various making methods namely slip casting, plastic making and semi-dry pressing.

The plasticity data is plotted on a clay workability chart (Fig.11) as used by the British Geological Survey (unpulished data). This chart shows how changes in plasticity index and plastic limit values of clays affect their usefulness for pottery and brick manufacture. Six samples (SS, U, M, NM, SM and S) show optimum properties with respect to plasticity, drying shrinkage, consistency and body strength. The rest of the samples show slightly higher drying shrinkage but they are still within the moderate range. Only one sample, that is SS, seems not suitable for pottery industry whereas the rest are well within the specifications required for pottery manufacturing. Eleven samples (NT, WA, N, AA, SU, CM, M, W, SS, NM and SM) fell in the area whereby clays are suitable for brick manufacturing.

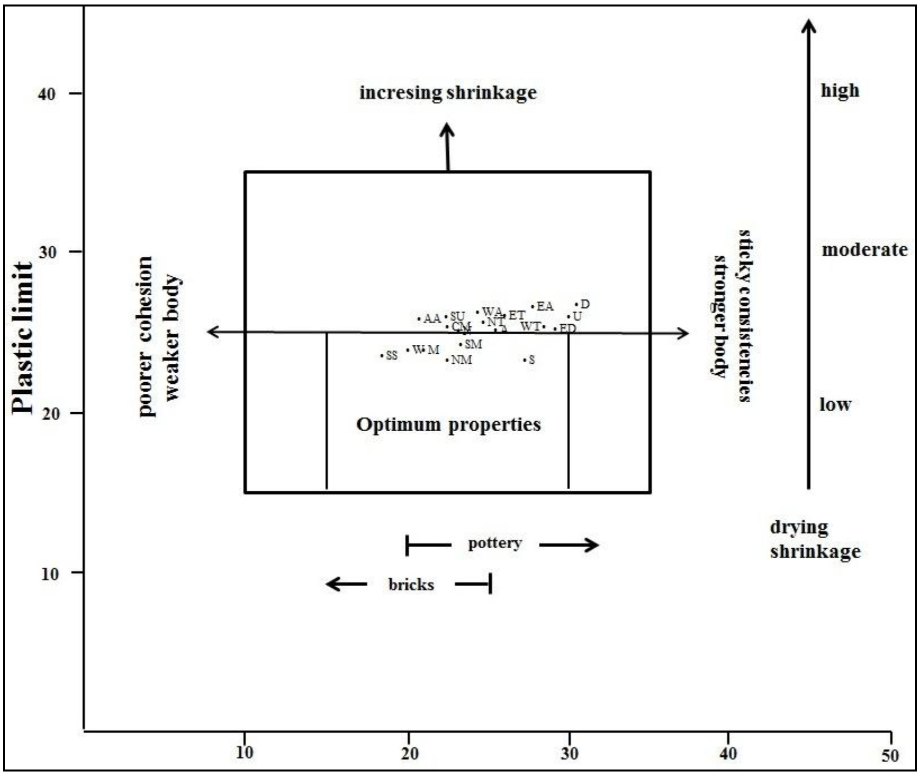

Fig.11: Clay workability chart (Atterberg limit of brick and pottery clays – Unpublished data from the British Geological Survey)

- **Refractory and fire-clay refractory bricks (chamotte i.e.-aluminosilicate)**

Kaolin clay is the main ingredient for manufacturing refractories (Abu-Hamatteh and Al-Taie, 2003). The grade of kaolin clay to be used is basically assessed on its chemical constituents especially alumina content and fluxing compounds (iron, alkalies and alkaline earths). Good refractory grade clay contains high alumina content (at least 30%) and very low fluxing elements (Ryan, 1978). Jindy and Ibrahim (1982) found that the pyrometric cone equivalent (P.C.E) of some of the white Ga'ara clays is higher than cone 26 (i.e. more than 1605 ºC).

Accordingly, nine of the investigated clay deposits are suitable for refractories; namely ET, EA, WT, A, ED, D, U, WA and S. The present study on the plasticity of the Ga'ara kaolin clays showed that they are plastic. Such excessive plasticity can be reduced by the addition of pre-calcined kaolin (grog) which is coarser in particle size. The addition of grog reduces plasticity and shrinkage and improves refractoriness. The addition of grog is well known way to manufacture chamotte bricks (Djangang *et al*., 2008, Trurong *et al*., 2018).

The suitability of the Ga'ara kaolin clay, particularly Duekhla deposit, for refractory industries was also assessed by previous workers such as Mustafa (1981); Jindy and Ibrahim (1982) and Ghanim (1996). However, the Duekhla Kaolin clay was used in the manufacturing of medium-duty aluminosilicate firebricks (chamotte bricks) by mixing it with grog fired at 1300 ºC (60% grog + 40% raw clay) then moulding it, with about 8% moisture, under pressure 250 kg/cm^3 and firing at temperature about 1350 ºC.

- **Extraction of alumina and aluminium compounds**

Several laboratory experiments and bench tests were carried out on the Ga'ara kaolin clay from the Nijili deposit to extract alumina and aluminium compounds, such as alum. The work incorporated two steps, the first to extract impure alumina and the second to purify it to a high grade (Hammodi and Abdulla, 1987a). The purified and calcined alumina had 99.8 wt.% Al_2O_3, 0.080 wt.% Na_2O, 0.027 wt.% Fe_2O_3 and < 0.006 wt.% TiO_2. Preliminary tests were also carried out for the production of alum from coloured kaolin clay. Samples were collected from Wadi Al-Nijili coloured kaolin deposit. Alum was successfully produced and its quality seems to fall between Egyptian and Japanese imported alums (Hammodi and Abdulla, 1987b).

- **Paint manufacturing**

Kaolin clay is used as a white pigment although less satisfactorily than others such as titanium oxide. It can also be used as an extender in order to reduce the cost and improve the performance of the product (Mustafa, 1981 and Murray, 2007). In paint industry, the particle size, colour (especially brightness), ease of defloculation and minimal content of soluble salts are the most important characteristics of the clay (Murray, 2007). The present study shows that samples from eight deposits are best suited for such industry: North Wadi Tayyarah (NT), East Wadi Tayyarah (ET), East Afaif (EA), West Wadi Tayyarah (WT), East Duekhla (ED), Duekhla (D), Ubairan (U) and Semhat (S). These clay deposits have good reflectivity and the particles finer than two microns range between 65.9% in Semhat deposit (S) to 82.9% in Duekhla deposit (D). The Semhat deposit is hence inferior in quality in comparison to Duekhla from point of view of fine particle content, but superior in colour properties.

CONCLUSIONS

The investigated deposit-representative samples are plastic sedimentary (transported) kaolin clay which resembles in many respects the British Ball Clay. The majority of the Ga'ara Formation kaolin clay deposits can be classed as high-grade kaolin clay which are suitable for use in the manufacture of some industries such as paper, paints, rubber, plastics, cosmetics, pharmaceuticals and some special clay ceramics (such as white tableware, electro-porcelain and bone china). Others can be used for the above industries with minimal processing. All studied samples are suitable for general clay ceramics and some of them are fat clays with low fluxing compounds which are suitable for refractories.

ACKNOWLEDGEMENTS

Thanks are due to the officers of the Iraq Geological Survey and the Director General for allowing access to some of their data on the Ga'ara kaolin clay, supplying the bulk samples and their chemical analyses. Our thanks go to Professor Dr. Radko Kühnel and Dr. Khaldoun Al-Bassam for their critical reading of the manuscript.

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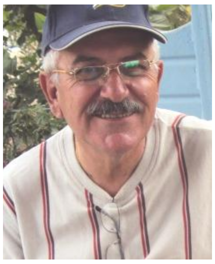

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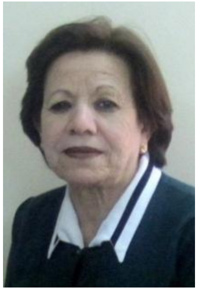

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