Science and Technology, 7 (2002) 259-277

Mineralogy of the Tertiary Clay Deposits in Makkah and Rabigh Quadrangles, West Central Arabian Shield, Saudi Arabia

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معدنية رواسب الحقب الثالث بمربعي مكة ورابغ بغرب وسط الدرع العربي ، المملكة العربية السعودية

محمد حسين بسيوني ، محمد أبو اليزيد العسكري ، نادى أديب سعد ، و رشدى جمال تاج

خلاصة: تمت دراسة التركيب المعدني لرواسب الطين بمربعي مكة ورابغ بواسطة جهاز حيود الاشعه السينية والتحليل الحراري التفاضلي والوزنى بالإضافة إلى طرق أخرى. وقد دلت النتائج على أن تلك الرواسب تتكون بصفة أساسيه من معدن مونتموريلونيت (النوع الغنى بالكالسيوم أو الماغنسيوم أو كلاهما) ، معدن كاولينيت بمصاحبة قليل من معدن اليت وندره من معدن كلوريت. كما سجل وجود طبقات مختلطة من مونتموريلونيت – اليت بعينتين فقط. ومن واقع الوفرة النسبية لهذه المعادن والتي تم الحصول عليها من تحاليل حيود الأشعة السينية أمكن تقسيم رواسب الطين محل الدراسة إلى ثلاثة أنواع : النوع الأول وهو الأكثر شيوعا غنى بمعدن مونتموريلونيت ، الثاني عباره عن خليط من معادن مونتموريلونيت يليه كاولينيت ثم اليت ، الثالث غنى بمعدن كاولينيت مع بعض مونتموريلونيت. وبصفه عامه لوحظ ازدياد معدن كاولينيت جنوبا بمربع مكة بينما اظهر معدن كلوريت – كمكون غير أساسي – ازدياداً فى اتجاه الشمال بمربع رابغ.

ABSTRACT: The mineralogy of the Tertiary clay deposits in Makkah and Rabigh quadrangles was thoroughly investigated by X-ray diffraction and differential thermal and thermogravimetric analyses in addition to other techniques. Results show that the investigated samples are predominantly composed of montmorillonite (Ca++ and/or Mg++ rich variety) and kaolinite, associated with subordinate illite and minor chlorite. Mixed layer montmorillonite-illite is recorded only in two samples. The relative abundance of these minerals by X-ray diffraction analysis showed that the studied clay deposits are of three types. The first, which is the most common, is highly montmorillonitic, the second is made up of a mixture of montmorillonite followed by kaolinite and illite and the third is highly kaolinitic with some montmorillonite. Generally, kaolinite shows a southward increase in Makkah quadrangle while chlorite, as a minor component, shows a northward increase in Rabigh quadrangle.

KEYWORDS: Mineralogy, Clay Deposits, Kaolinite, Montmorillonite-Illite and Makkah Quadrangle.

1. Introduction

Makkah and Rabigh quadrangles are located in the west-central part of the Arabian Shied bordering the Red Sea between Latitude 21° 00' and 23 ° 00' N. and Longitude 38° 50' and 40° 30' E. Both quadrangles comprise Precambrian rocks unconformably overlain by Tertiary sedimentary rocks in the west, and by Miocene to Pliocene lavas in the north (Moore and Al-Rehaili, 1989). There are extensive areas of Quaternary surficial deposits (sand and gravel) on the coastal plain and in the major wadies (Figure 1).

The Tertiary stratigraphic succession is subdivided by Moore and Al-Rehaili (1989) into the following formations starting from the oldest : Haddat Ash Sham, Usfan, Shumaysi, Khulays, Buraykah, Ubhur and Daffin. The latter is exposed only in Rabigh quadrangle. Lithologically all these formations consist mainly of siliciclastic rocks (sandstones, siltstones, clays, conglomerates, etc.) presumably derived from the surrounding Precambrian basement and pre-existing sedimentary rocks.

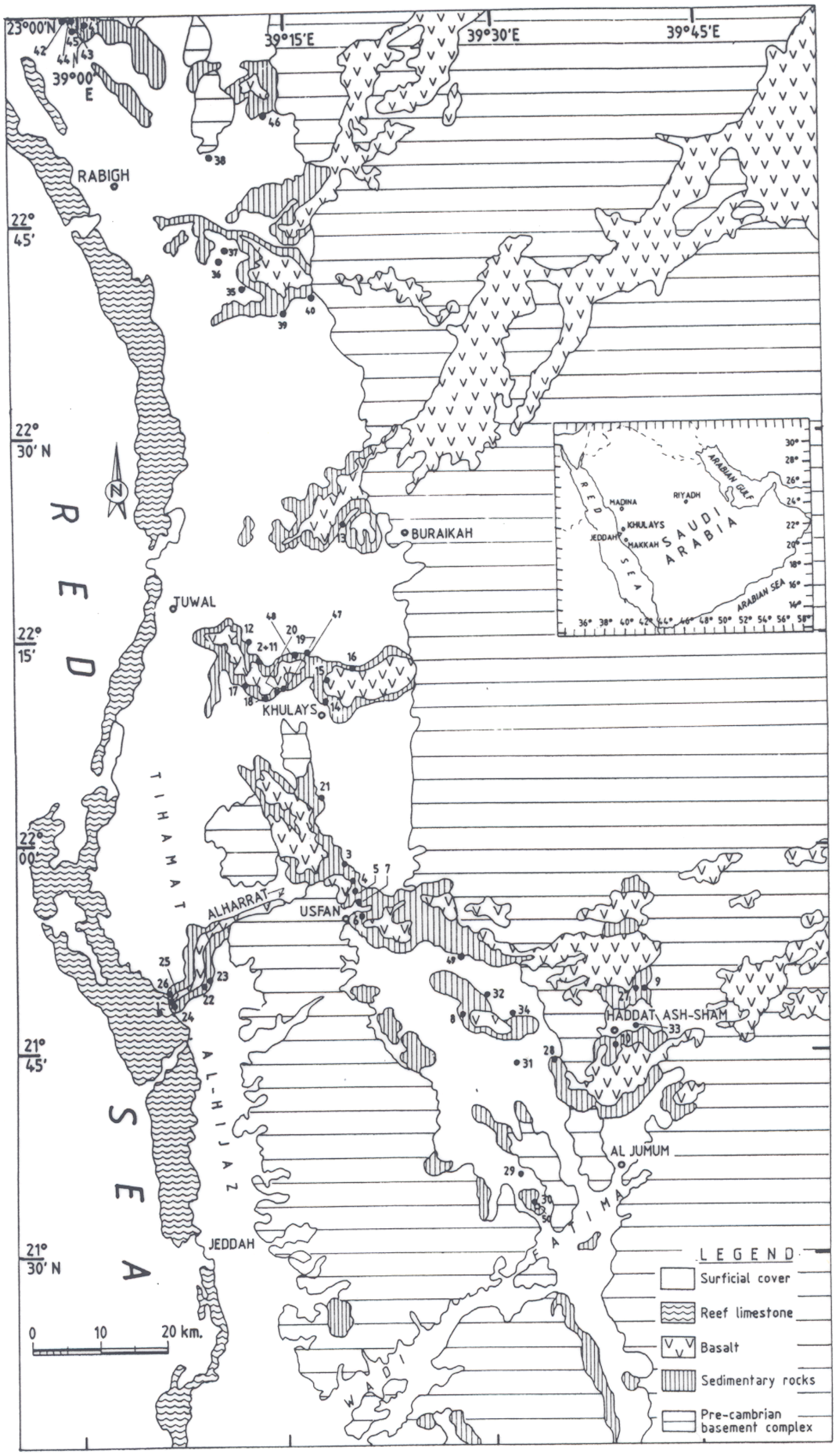

Figure 1. Simplified geological and location map of the studied stations in the Makkah and Rabigh quadrangles (Stations 1, 22, 23, and 24 are located in Ubhur Formation. and studied by Taj *et al*., 2002 b).

However, the overwhelming clastic sedimentation is temporarily interrupted by a limited but a persistent marine and lacustrine carbonate units in Usfan, Ubhur and Daffin formations and evaporites (mainly gypsum) in the last two formations. Clay deposits of all the formations are usually capped with basaltic flows called Harrat.

The geology, mineralogy and chemistry of all these Tertiary formations have been thoroughly investigated by Taj *et al*. (2001a). The economic potential for possible use of these clay deposits in different fields have been evaluated by Taj *et al*. (2001b). Based on geochemical evidence, Taj *et al*. (2002a) concluded that the Tertiary clay deposits have been deposited under transitional to marine environments. The mineralogy and sedimentary phenomena characterizing the clay (spheroids) and evaporite (desert roses) deposits of Ubhur Formation have been studied by Taj *et al*. (2002b). The mineralogy of the clay samples collected from forty six stations located in Makkah and Rabigh quadrangles (Figure1) and belonging to the other Tertiary formations is the subject of the present study. Some representative samples have been subjected to grain size analysis.

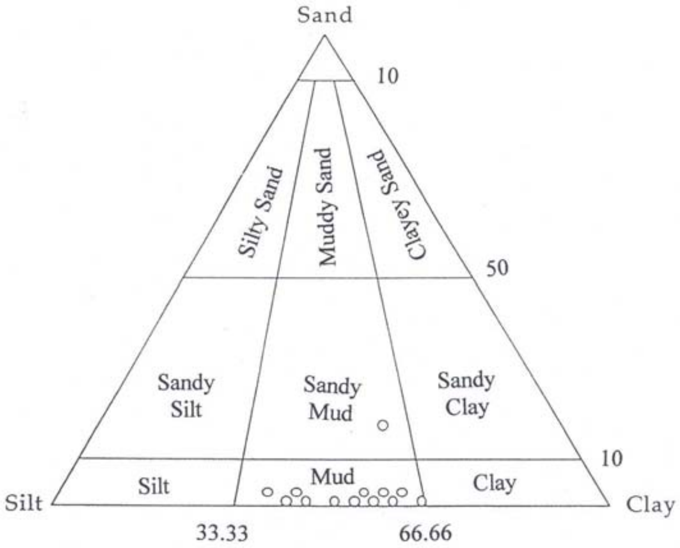

Figure 2A. Plot of the analyzed samples on the (Sand-Silt-Clay) ternary diagram of Folk (1980).

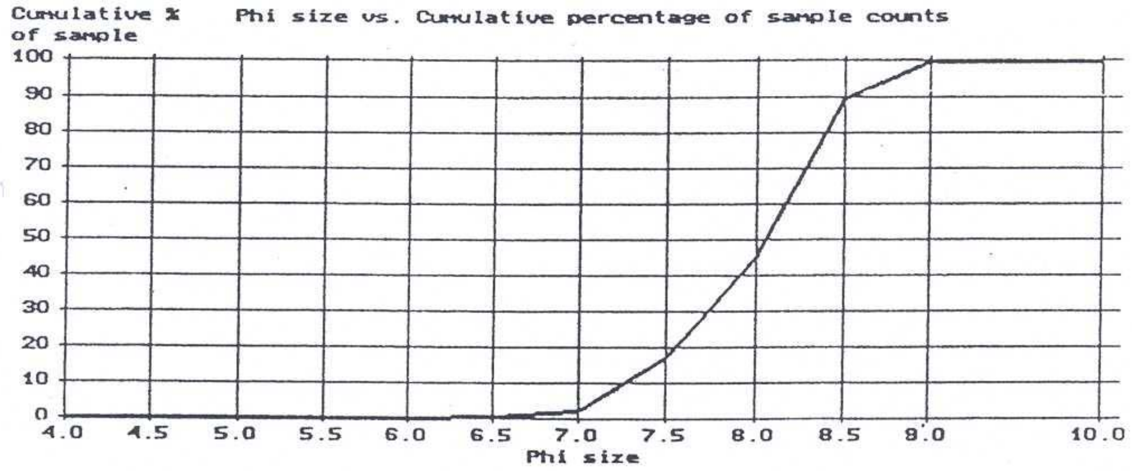

Figure 2B. Average phi size vervus cumulative percentages of the analyzed samples using Laser Particle Counter.

2. Grain Size Analysis

Grain size analysis has been performed for thirteen selected clayey samples representing the studied formations. Sand fraction is determined by wet sieving while the silt and clay fractions are determined using Laser Particle Counter. The results are plotted on Folk's (1980) ternary diagram (Figure 2A) which shows that 12 samples are located in the mud field and only one sample belongs to sandy mud. Plots of the mud samples are located along the clay-silt base but more nearer to the clay apex indicating the dominance of clay fraction in most samples. The sandy mud sample is also characterized by the dominance of clay fraction over sand and silt. The average cumulative curve constructed for the studied samples (Figure 2B) shows that the wide majority of the phi sizes are restricted to 7 – 9 ϕ with higher concentration in 8 ϕ (Md 8.05 ϕ ; Mz 7.98 ϕ). Values of the other grain size parameters, calculated using the formulae of Folk and Ward (1957), show that the studied samples are generally well sorted, strongly fine-skewed and meso-to leptokurtic.

3. Mineralogy

The mineralogy of the clay deposits of the different Tertiary formations is thoroughly investigated by X-ray diffraction, using a PW 1840 Philips diffractometer, and differential thermal and thermogravimetric analyses using the computerized Perkin Elmer DTA 7 and TGA 7 units. Some samples are also examined by Zeiss scanning and transmitted electron microscope and by a Perkin-Elmer infrared spectrophotometer, Model 1430 (Central Laboratory, Faculty of Science, Alexandria Univesity). Some staining tests are also performed.

3.1 X-Ray Diffraction Analysis

The clay fraction of eighty nine surface samples representing the studied formations and nine subsurface clay samples from different depths at different locations have been analyzed by X-ray diffraction. In addition, eight representative bulk samples have been analyzed for the associated non-clay minerals. Identification of clay minerals is based on the data given by Brown and Brindley (1980) and Powder Diffraction File, JCPDS (1984). Clay minerals are semiquantitatively estimated using the method given by Pierce and Siegel (1969).

Results of the X-ray diffraction analysis show that the clay samples under investigation are predominantly composed of montmorillonite (Ca++ and/or Mg++ rich variety) and kaolinite, associated with subordinate illite and minor chlorite. Mixed layer montmorillonite-illite is recorded only in two samples; one belongs to Haddat Ash Sham Formation and the other is a subsurface sample. Diffractograms of representative samples from the studied formations are given in Figure3, while the distribution and relative abundance of the identified minerals are given in Table 1.

Some of the investigated clay samples are more or less monomineralic; formed of either montmorillonite as in Khulays samples from stations KH2, 3 and 4 ; or kaolinite as in Shumaysi samples from station SH 30 (Table 1). Both minerals especially kaolinite occurs in a highly crystalline state as indicated by the great sharpness of its characteristic peaks (Figure 3).

X-ray diffractograms shown in figure 4 are for three representative bulk clay samples in which quartz followed by k. feldspar, plag. feldspar and alunite are the associated non-clay minerals.

3.2 Differential Thermal and Thermogravimetric Analyses

The clay fraction of seven clay samples representing some of the studied formations and one of the subsurface samples have been selected for the differential thermal and thermogravimetric analyses. Heating was up to 1050° C with a heating rate of 10^o C per minute. The results obtained are given in Table 2, which also shows the qualitatively identified clay minerals arranged in a decreasing order of abundance. Figure 5 shows the DTA, TG and DTG curves of four representative samples.

Thermal analyses results, given in Table 2 and illustrated in Figure 5, show the presence of three thermal effects. The first is represented by endothermic DTA peak below 200° C attended by endothermic DTG peak at about 90° C and a substantial TG loss of mass due to the removal of adsorbed water ranging from 3.3 % to 9.5 % of the original sample weight. The DTA peak is doubled in samples KH3, KHF1, 10, 31, 65 and 79 (Table 2 and Figure 5) indicating not only the predominance of montmorillonite, but also its variety with Ca2+ , Mg2+ and Ba2+ as exchange cations (Todor, 1976).

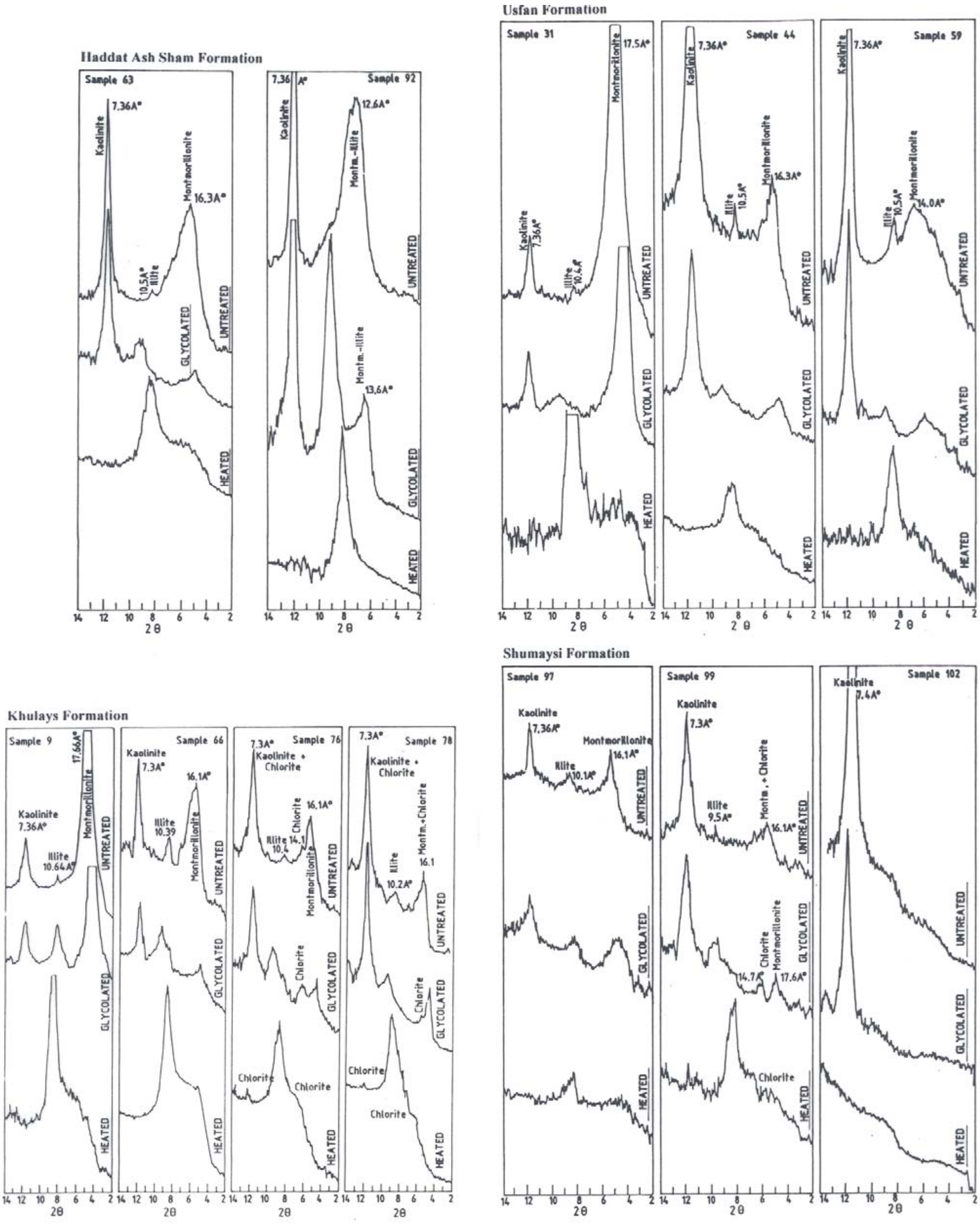

Figure 3. See next page for legend.

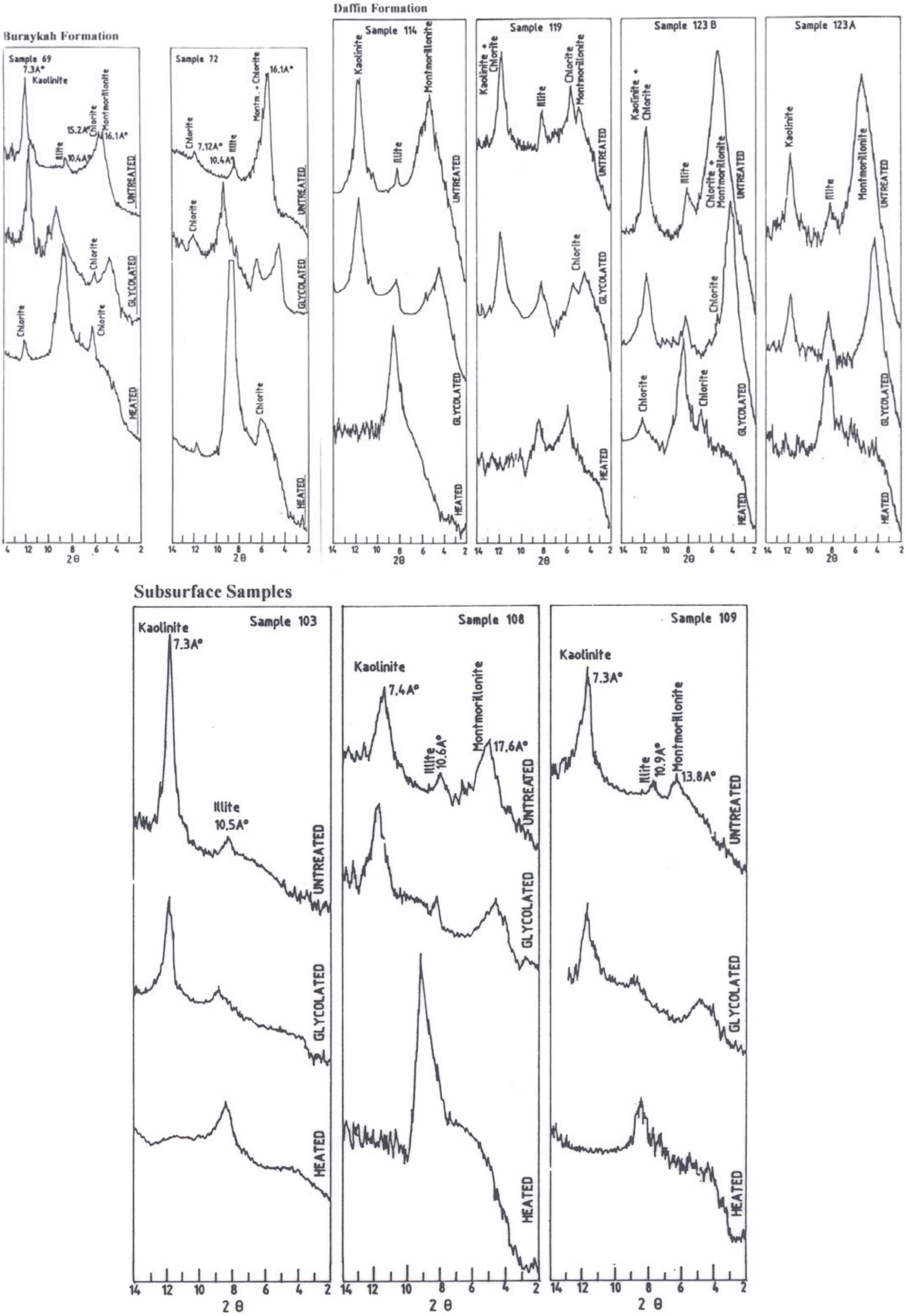

Figure 3. X-ray diffraction patterns of the clay fraction (< 2 μ m) of representative samples from the investigated clays.

Table 1: Identified clay minerals and their relative abundances of the studied samples (clay fraction), based on X-ray diffraction analysis.

Formation	Station No.	Sample No.	Montmor-illonite (M)	Kaolinite (K)	Illite (I)	Chlorite (Ch)	Mixed Layer (M-I)	Remarks
Haddat Ash Sham	HS10	62	++	+++	+++	-	-	
		63	++++	+++	++	-	-	
		64	+++	+++	++	-	-	
	HS27	92	-	++	-	-	++++	
							+	
Usfan	US6	26	+++++	++	+	-	-	
	US7	28	++++	+++	+	+	-	
		30	+++++	++	+	+	-	
	US8	31	+++++	+	+	-	-	
		32	++++	+++	++	-	-	
		33	++++	++	+	-	-	
		34	++	++++	+	-	-	
		36	++	+++++	+	-	-	
		38	+++	++++	++	-	-	
		39	++	++++	++	-	-	
		40	+++++	++	-	-	-	
		41	++++	+++	-	-	-	
		42	+++	+++	++	-	-	
		43	+++++	+	+	-	-	
		44	++	++++	++	-	-	
		45	++++	++	+	-	-	
		46	+++	+++	++	-	-	
		47	+++++	++	+	+	-	
		49	++++	++	++	-	-	
		50	++++	++	++	-	-	
		52	+++	++++	++	-	-	
		53	+++	++++	++	-	-	
		53A	+++	+++	++	-	-	
		54	+++	++++	-	-	-	
		56	+++	++++	-	-	-	
		58	+++	+++	++	-	-	
	US9	59	++	++++	++	-	-	
		61	+++	++++	-	-	-	
Shumaysi	SH30	97	++++	++	++	-	-	
		98	++++	+++	++	-	-	
		99	++	++++	++	+	-	
		100	-	+++++	-	-	-	
		101	+++++	+	+	-	-	
		102	-	+	-	-	-	
				+++++				
				+				
	SH50	132	++++		++	-	-	
		133	++++		+	-	-	
		134	+++++	++	+	-	-	
		135	++	++	+	-	-	
				++				
				++++				

Table 1: (Continuous)

Formation	Station No.	Sample No.	Montmor-illonite (M)	Kaolinite (K)	Illite (I)	Chlorite (Ch)	Mixed Layer (M-I)	Remarks
Daffin		114						
	DF37	115	+++	+++	++	-	-	
	DF38	116	+++	+++	++	-	-	
	DF40	117	+++	++	-	+++	-	Swelling chlorite
		118						
	DF41		++	++	++	++	-	
			++	+++	++	++	-	
		119						
	DF42	121	++	+++	++	++	-	
	DF43	122	-	+++	++	+++	-	
	DF44	123A	+++	+++	++	++	-	
		123B						
	DF45		++++	++	++	-	-	
			+++	++	++	++	-	Core of spheroids
		125						
	DF46		+++	++	-	+++	-	Spalls of spheroids
								Swelling chlorite
Sub-surface		103				-		
		104				+		
	31		-	++++++	+		-	
		105	+++	+++	++	+	-	
	32		+++++	++	+		-	
		106				+		
		107	++++	++		-		
	33	108	+++++	++	++	-	-	
			++	+++	+		-	
		109			++	-	-	
		110	-			-		
	34	111	++++	+++++	+	-	++	
			++++	+++	-		-	
				+++	++		-	
					+			

Symbols for approximate percentage contents are ++++++:more than 90;+++++:70-90;++++:50-70;+++:30-50; ++:10-30;and +lessthan10;-:not recorded.

Table 1: (Continuous)

Formation	Station No.	Sample No.	Montmor-illonite (M)	Kaolinite (k)	Illite (I)	Chlorite (Ch)	Mixed Layer (M-I)	Remarks
Khulays	KH2	KH1	+++++	+	+	-	-	
		KH2	+++++	+	+	-	-	
		KH3	++++++	+	+	-	-	
		KH	++++++	+	+	-	-	Composite sample (kH1,2,3)
	KH3	KHF1	++++++	+	+	-	-	
		KHF2	++++++	+	+	-	-	
		KHF3	+++++	++	++	+	-	
		KHF	++++++	+	+	-	-	
		7	+++++	+	+	-	-	
		8	+++++	++	+	-	-	Composite sample (kHF1,2,3)
		9	++++++	+	+	-	-	
		10	++++++	+	+	-	-	
	KH4	11	++++++	+	+	-	-	
		14	+++++	+	+	+	-	
		17	+++++	+	+	-	-	
	KH5	18	+++++	++	+	-	-	
		19	+++++	++	+	-	-	
		20	+++++	++	+	-	-	
		22	+++++	++	+	+	-	
		23	+++++	++	+	+	-	
		25	+++++	++	-	+	-	
	KH11	65	+++	+++	+++	-	-	
		66	+++	+++	+++	-	-	
		67	++	+++	+++	-	-	
	KH12	68	++	+++	++	++	-	
	KH14	74	++	++++	++	+	-	
		75	++	+++	+++	++	-	
		76	++	++++	++	++	-	
		77	++++	++	++	-	-	
		78	++	+++++	+	+	-	
		79	++++++	+	+	-	-	
		80	++	++++	++	-	-	
	KH16	82	++++	++	+	+	-	
	KH17	83	+++	++	++	-	-	
		84	++++	++	++	+	-	
	KH20	87	++	++++	++	+	-	
	KH47	126	++++	++	++	-	-	
	KH49	131	+++	++++	-	-	-	
Buraykah	BR13	69	+++	+++	++	+	-	
		70	++++	+++	-	+	-	
		71	++++++	-	+	-	-	
		72	+++++	-	++	++	-	
		73	+++++	++	++	+	-	

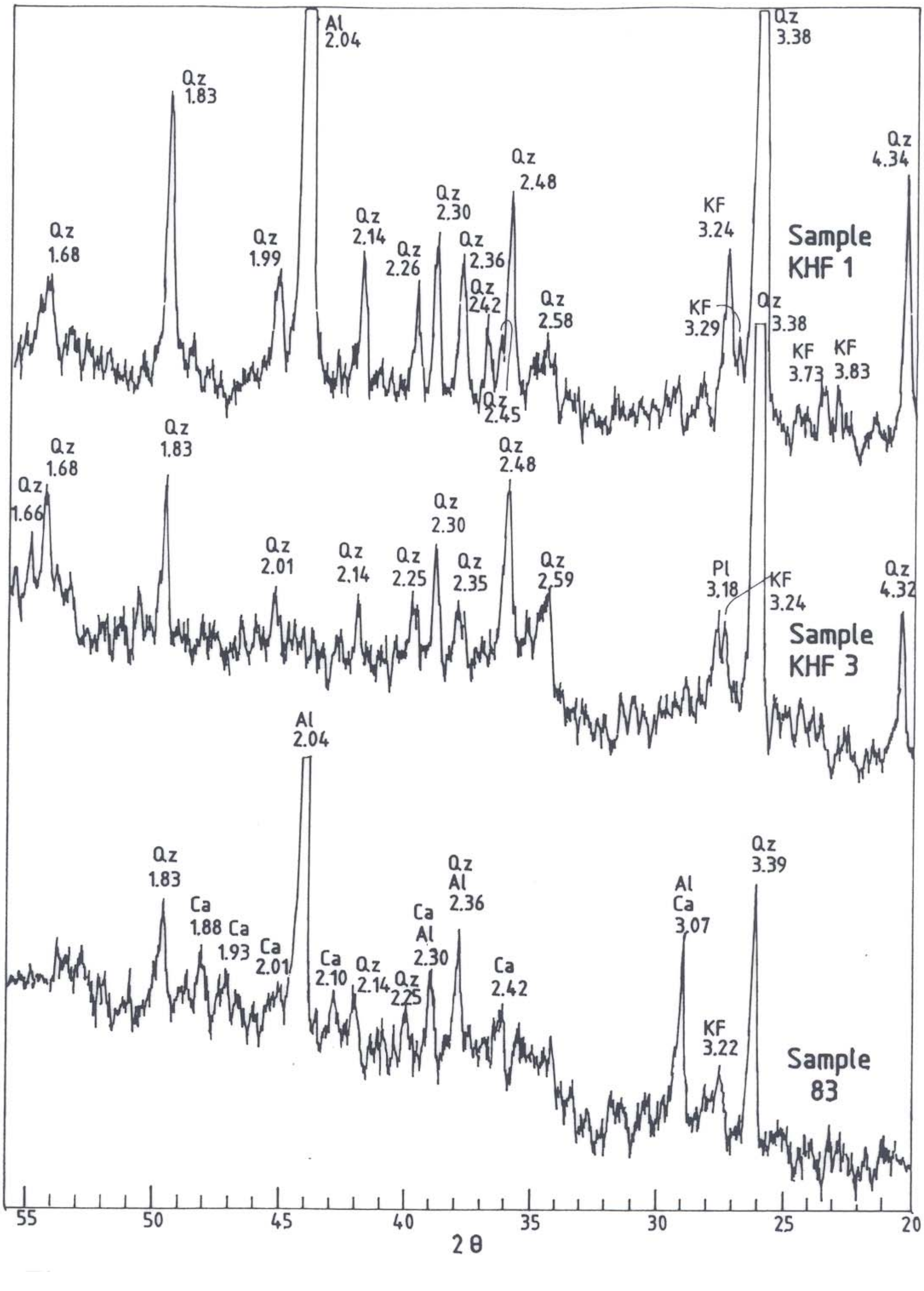

Figure 4. X-ray diffraction patterns of representative bulk samples of the investigated clays. Qz-Quartz, KF-Potash Feldspar (mainly microcline), Pl-Plagioclase Feldspar, Ca-Calcite and Al-Alunite.

The varying TG percentage loss indicates the varying amounts of montmorillonite (Table 2). The highst TG percentage loss (9.5% %) in samples KH3, 31 and 79 indicates the highst montmorillonite content in these samples. On the other hand, the relatively lower percentage loss (8.1 %), exhibited in sample 65 (Figure 5) , indicates the presence of other minerals having lower adsorbed water content like kaolinite and illite as less common minerals. In samples 102 (Figure 5) and 103 (Table 2), the DTA peak is smaller and not doubled and the TG percentage loss are too small (3.3 % and 3.6 %) suggesting the presence of weakly crystalline kaolinite as a major constituent.

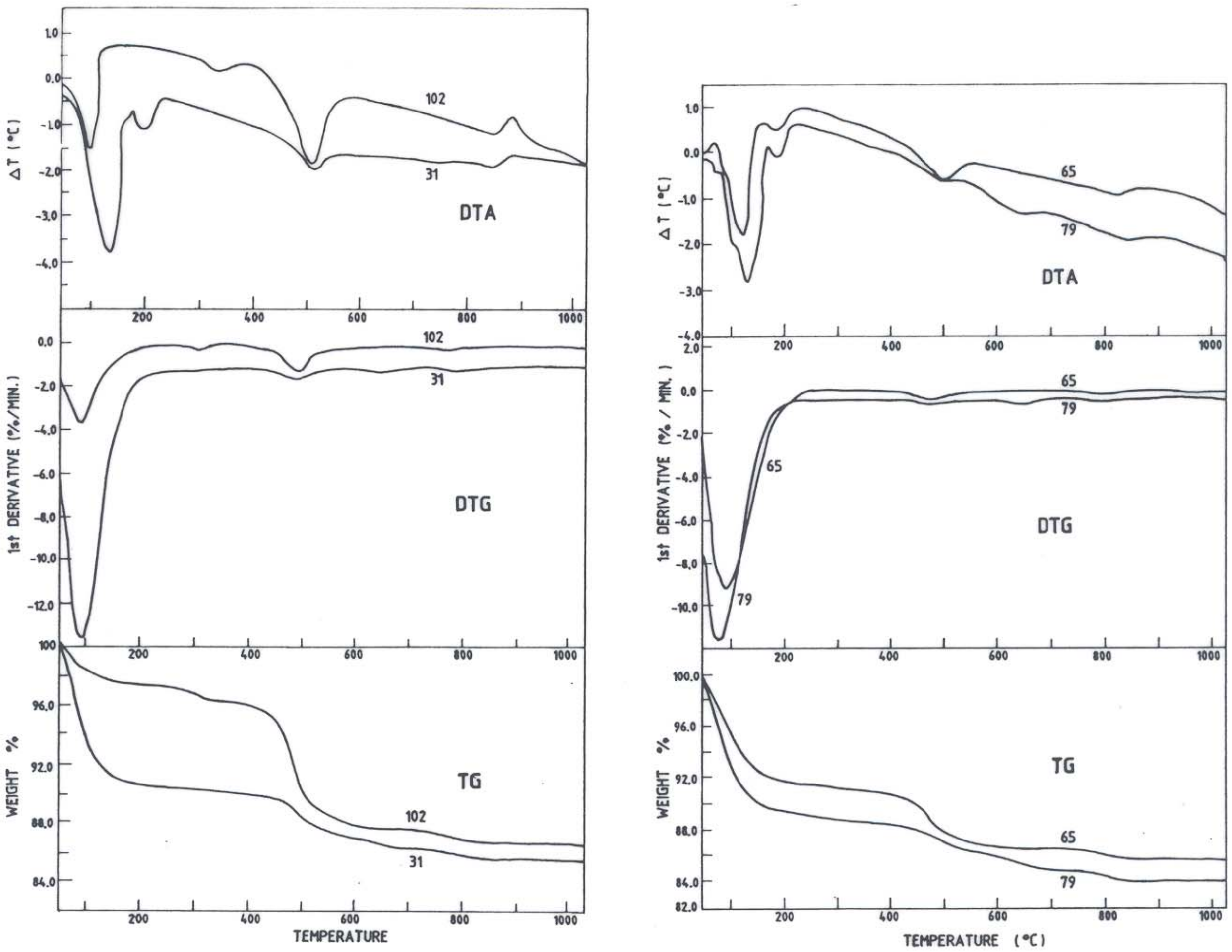

Figure 5. Differential thermal and thermogravimetric curves of four representative clay samples (sample 31 from Usfan Formation, sample 102 from Shumaysi Formation and samples 65 and 79 from Khulays Fprmation).

Sample 102 shows an endothermic DTA peak at 330 °C accompanied by a similar DTG peak at 305 °C and a TG loss of 1.1 % due to the removal of water from goethite (Table 2).

The second thermal effect is also endothermic and represented by DTA peak at 513° C to 539° C attended by a similar, but smaller, DTG peak at 471° C to 488° C, and a loss of mass due to the loss of hydroxyl group, ranging from 2.2 % to 6 % (in samples KH3, KHF 1, 10, 31, 65 and 79). These values confirm the dominance of montmorillonite with lesser amounts of kaolinite and illite (Table 2 and Figure 5). The highst TG percentage loss values exhibited in samples 102 (8.3 %) and 103 (8.9%), confirm the dominance of kaolinite (Table 2 and Figure 5).

This thermal effect is followed by another endothermic effect, but of lesser extent, represented by DTA peak at 841° C to 880° C and DTG peak at 780° C to 818° C and minor TG percentage loss ranging from 0.6 % to 1.2 %. This loss of mass is due to the dehydroxylation, i.e., the removal of OH group from the lattice of clay minerals (especially montmorillonite, illite and chlorite) which had not been fully removed. Sample 79 shows an additional dehydroxylation endothermic effect represented by a minor DTA peak at 665° C attended by minor DTG peak at 635° C and a TG loss of 1.9 %. This effect, most probably, indicates the presence of minor chlorite (Table 2).

Table 2: Differential thermal and thermogravimetric analyses results of the investigated clay samples.

Formation and Station No.	**Sample No.**	**Endoth.peak**		**Exoth. DTA**	**TG loss**	**Description of DTA peak**		**Assignment of the reaction**	**Identified minerals**
		DTA °C	**DTG °C**	**Peak °C**	**%**				
Khulays	**KH3**	129	90	-	9.5	double	large, sharp	removal of adsorbed water.	mainly montmorillonite, probably with minor amounts of illite + kaolinite
		186					small		
(2)		520	475	-	4.9	mod. slightly asym.		dehydroxylation, i.e.	
		851	818	-	1.2	small, broad		removal of OH group.	
		-	-	852	-	very small, sym.		structural reorganization.	
Khulays	**KHF1**	125	82	-	9.1	double	large, sharp	removal of adsorbed water.	mainly montmorillonite, probably with minor amount of kaolinite + illite.
		193					small		
(3)		513	486	-	4.9	mod., slightly asym.		dehydroxylation, i.e.	
		855	786	-	1.0	very small, broad		removal of OH group.	
		-	-	894	-	very small, sym.		structural reorganization.	
Shumaysi	**10**	125	79	-	8.2	double	large, sharp	removal of adsorbed water.	mainly montmorillonite, probably with minor amount of kaolinite + illite.
		199					small		
(3)		539	484	-	6.0	mod., silghtly asym.		dehydroxylation, i.e.	
		862	780	-	0.6	small, broad.		removal of OH group.	
		-	-	903	-	very small, sym.		structural reorganization.	
Usfan	**31**	130	78	-	9.5	double	large, sharp	removal of adsorbed water.	mainly montmorillonite, probably with minor amount of kaolinite + illite.
		194					small		
(8)		523	482	-	3.3	mod., slightly asym.		dehydroxylation, i.e.	
		670	640	-	1.5	small, broad		removal of OH group.	
		861	781	-	0.7	small, broad			
		-	-	900	-	very small, sym.		structural reorganization.	

The last thermal effect is exothermic and immediately follows the dehydroxylation effects. It is represented by a small to moderate DTA peak between 852° C and 909° C which is not accompanied by any weight change. This exothermic reaction is attributed either to structural reorganization or recrystallization, or to the formation of high-temperature mineral phases like mullite, cristobalite, etc. This exothermic DTA peak is generally small to very small in all samples except samples 102 (Figure 5) and 103 (Table 2) in which it is relatively larger confirming the presence of kaolinite as a major constituent.

Ten representative bulk samples from different formations have been investigated thermogravimetrically using a Mettler TA 3000 apparatus. TG and DTG curves of four of these samples are shown in Figure 6 and their thermal interpretations are given in Table 3. Based on these data, the identified minerals and their order of abundance agree with the results arrived at by using the Perkin-Elmer TGA unit.

The quantitative composition of particular clay minerals cannot be determined because of the complex mineral composition of the studied samples. The thermal analyses provided the qualitative determination of the existing clay minerals and which of them are dominant in the samples and the results are in accordance with those of XRD.

Table 3: Thermogravimetric analyses results of some representative bulk samples from Shumaysi and Daffin formations.

Formation and Station No.	Sample No.	Endothermic DTG peak °C	TG loss %	Description of DTG peak	Assignment of the reaction	Identified minerals
Daffin	**119**	77	4.86	large, sharp, sym.	removal of adsorbed water.	kaolinite, montmorillonite
(42)		484 650 & 790	7.93	moderate, slightly asym. very minor.	dehydroxylation, i.e. removal of OH group.	with some chlorite and illite.
Shumaysi	**132**	101	6.84	large, sharp, slightly asym.	removal of adsorbed water.	montmorillonite, kaolinite,
(50)		504 640	5.49	moderate, sym. very minor.	dehydroxylation, i.e. removal of OH group.	
Shumaysi	**134**	80	2.61	mod. to large, sharp, sym.	removal of adsorbed water	kaolinite, montmorillonite,
		298	0.82	small, asym.	removal of water from hyd.Fe2O3	with minor illite and goethite.
(50)		502	6.07	large, sharp, sym.	dehydroxylation, i.e. removal of OH group.	
Shumaysi	**135**	83	6.66	mod. to large, sharp, sym.	removal of adsorbed water	montmorillonite, kaolinite,
		307	0.99	small, asym.	removal of water from hyd.Fe2O3	
(50)		518 660 & 780	5.17	large, sharp, sym. very minor.	dehydroxylation, i.e. removal of OH group.	

In case of the clay deposit that contain only one mineral, it is easy to make quantitative estimation for this mineral (El Askary, 1983). However, in clay mixtures, Grim (1968) showed that there are many complications, which inhibit quantitative estimation of clay minerals.

3.3 Scanning and Transmitted Electron Microscope

Five representative clay samples were investigated by the scanning electron microphotographs (SEM), out of which three were also investigated by the transmitted electron microphotographs (TEM). The study reveals the presence of the same clay minerals already identified by the other techniques. Identification is based on comparison with SEM given by Welton (1950) and given by Beutelspacher and Van Der Marel (1968). The study reveals the presence of montmorillonite, kaolinite and illite and their relative abundance are in agreement with the other studied techniques. SEM and TEM photographs given in plate 1 are selected to show these minerals.

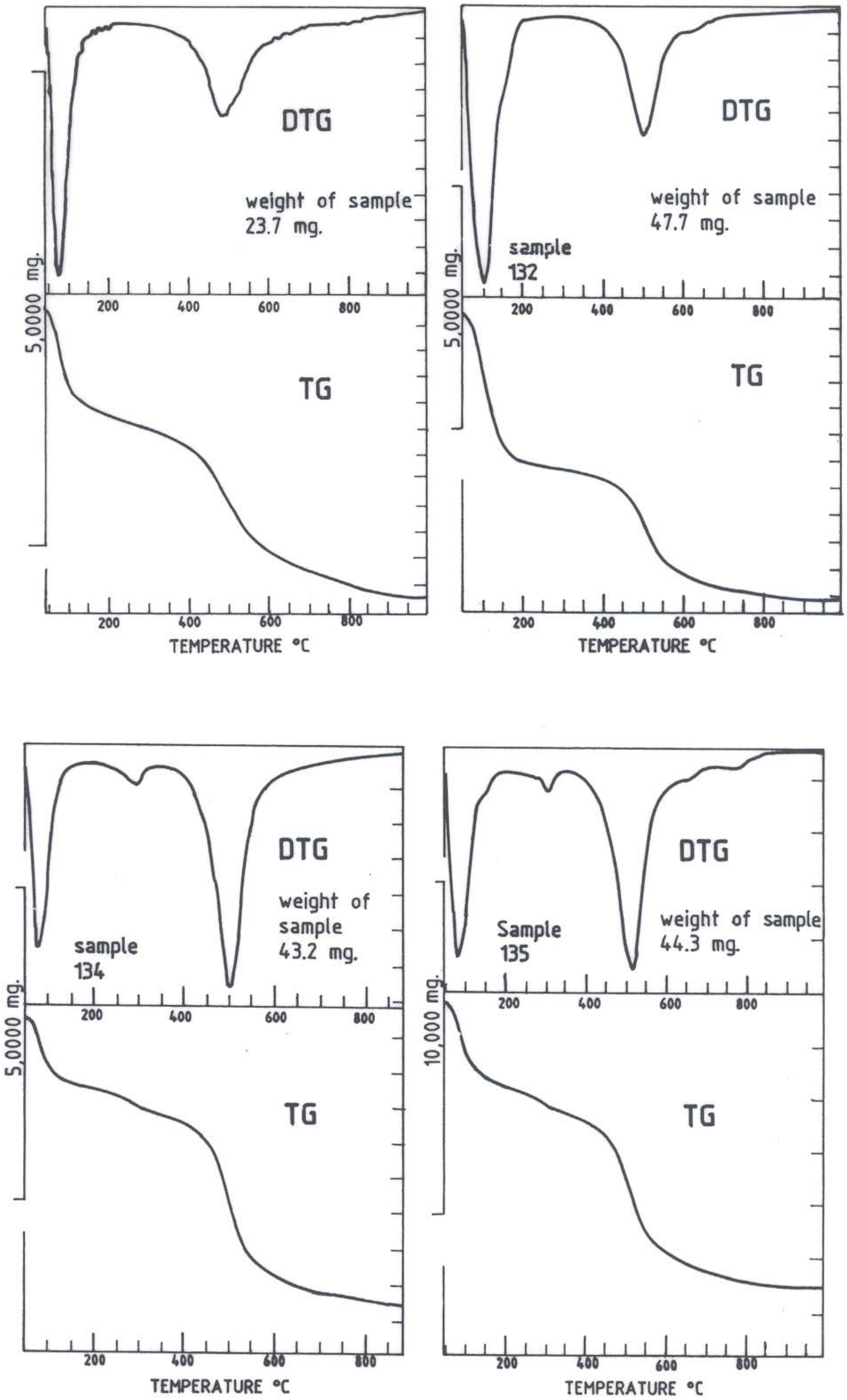

Figure 6. Thermogravimetric and differential thermogravimetric analyses curves of four representative bulk clay samples.

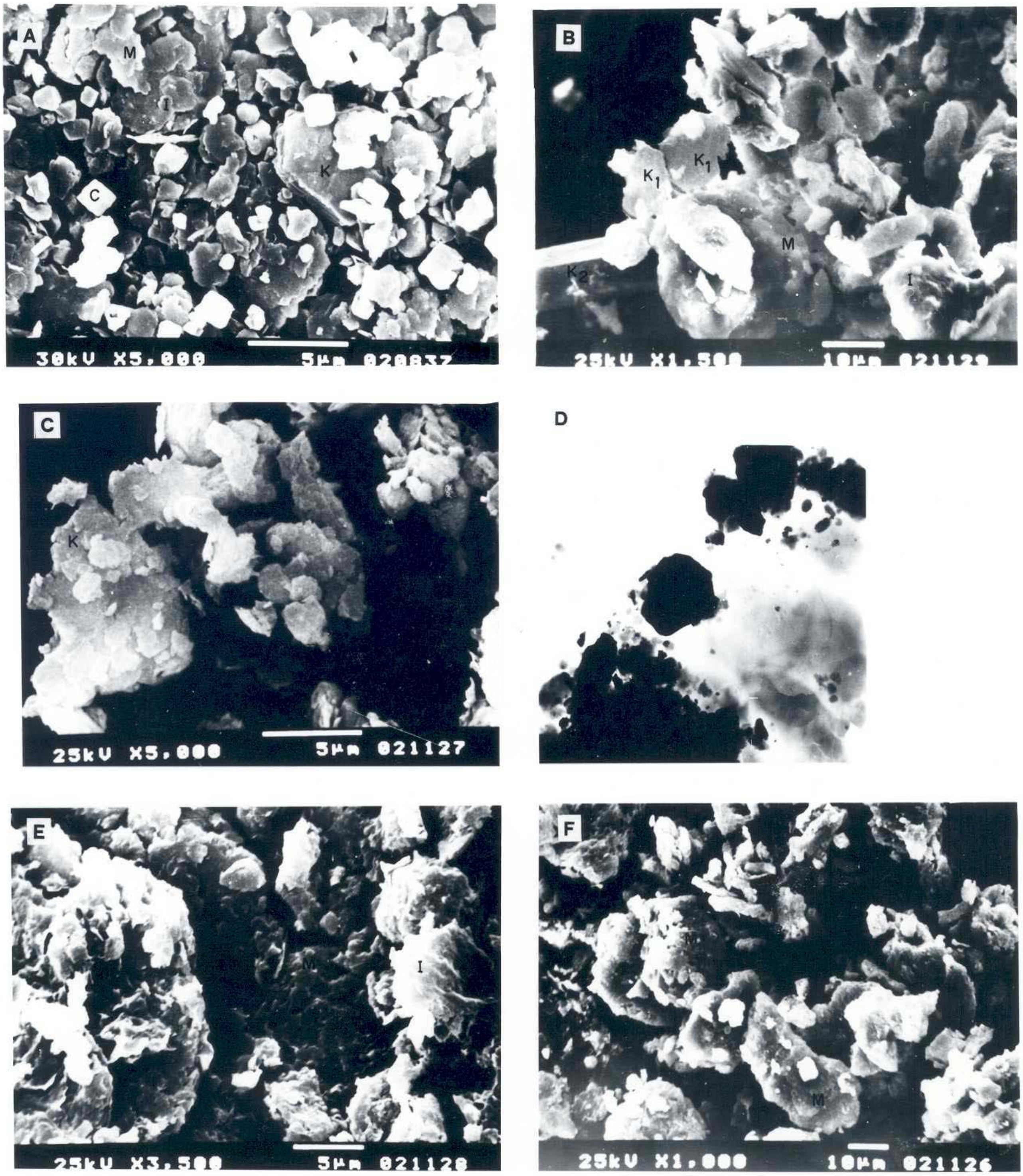

Plate I:

A- SEM photograph of sample 59 (Haddat Ash Sham Formation) showing pseudohexagonal kaolinite flakes (K) of different sizes with some ragged-edged, crenulated montmorillonite particles (M) and similar thicker illite particles (I). Minor calcite rhombs (C) are also seen.

B - SEM photograph of sample 69 (Buraykah Formation) showing poorly developed six-sided kaolinite flakes (K1) with crenulated montmorillonite particles (M) and similar thicker illite particles (I). An individual elongate kaolinite “verm” (K2) is also seen.

C - SEM photograph of sample 102 (Shumaysi Formation) showing poorly crystalline kaolinite flakes (K). The six-sided outline is only crudely shown.
D - TEM photograph of the same sample (102) in which kaolinite flakes are more or less hexagonal (X 12, 500).
E - SEM photograph of sample KH3 (Khulays Formation) showing crenulated montmorillonite aggregates (M) with minor thicker flaky illite (I).
F - SEM photograph of sample 31(Usfan Formation) showing crenulated irregular montmorillonite aggregates (M).

4. Kaolinite

Kaolinite crystals are of different sizes and range from poorly to moderately crystalline. The moderately-well crystalline variety appears as pseudohexagonal flakes or plates with ragged edges (Plate 1a, sample 59). The sharpest XRD peaks of kaolinite are well recorded in this sample. Sample 69 (Plate 1b) shows individual kaolinite books arranged face- to – face into an elongate stack, called a “ verm” (Welton, 1950). The poorly crystalline variety shows less distinct development of hexagonal outlines. Crystal edges are very ragged and irregular and the hexagonal outline is only crudely shown (Plate 1c, sample 102). Transmitted electron microphotograph (Plate 1d) of the same sample shows irregular to poorly developed hexagonal outline.

5. Montmorillonite

Montmorillonite is present as ragged-edged and crenulated particles, some of them show flaky morphology (Plate 1e and f, samples KH3 and 31). The flake-shaped aggregates appear to be stackings of units without regular outline. They are discernible but frequently too small to be seen individually.

6. Illite

Illite is present as small poorly defined flakes commonly grouped together in irregular aggregates. Generally it resembles montmorillonite, but the particles are larger and thicker and have better-defined edges (Plate 1a, b and e.).

6.1 Infrared Analysis

Infrared absorption has been used extensively and successfully in recent years in mineral identification, especially clay minerals. The clay fractions of thirty nine samples representing the studied formations and those from the subsurface were investigated.

Many of the known mineral absorption bands have been published by many authors. Using the mineral absorption bands given by Moenke (1966), Lyon (1967), Farmer (1975), White (1975), Van Der Marel and Beutelspacher (1976) and Estep (1977) the main clay minerals forming the investigated clay samples were identified (Figure 7). Clay minerals identified in sample 89 A (Figure 7) which belong to Ubhur Formation agree with data of other techniques given by Taj *et al.* (2002 b). Generally, the qualitative results of infrared spectroscopy agree with other mineralogical investigations. The relative intensities of peaks are not a measure of the amounts of their corresponding minerals.

6.2 Staining Tests

In the present work, five representative samples varying in their clay-mineral composition were selected for staining tests with benzidine and malachite green dyes. Samples principally composed of montmorillonite e.g. KH3 and 31, showed color change to purple-blue with benzidine and yellow-red yellow with malachite green. The intensity of these colors increases with the increase of montmorillonite content and decreases, or even to great extent masked in the chocolate brown sample (KH3).

On the other hand, samples with high kaolinite content, e.g., 100 and 102 (Shumaysi Formation) showed no reaction with benzidine and sample 100 showed color change to bluish green with malachite green and the clay particles showed weak pleochroism when examined with the polarizing microscope. Sample 102 neither showed color change with malachite green nor gave any pleochroism because of its brownish color due to its high iron oxide content. Illite shows no reaction with benzidine and gave green blue-green with malachite green (Mielenz *et al*., 1950). Its detection in the investigated samples is difficult due to its relatively lower content and the masking effect exerted by the accompanying dominating clay minerals.

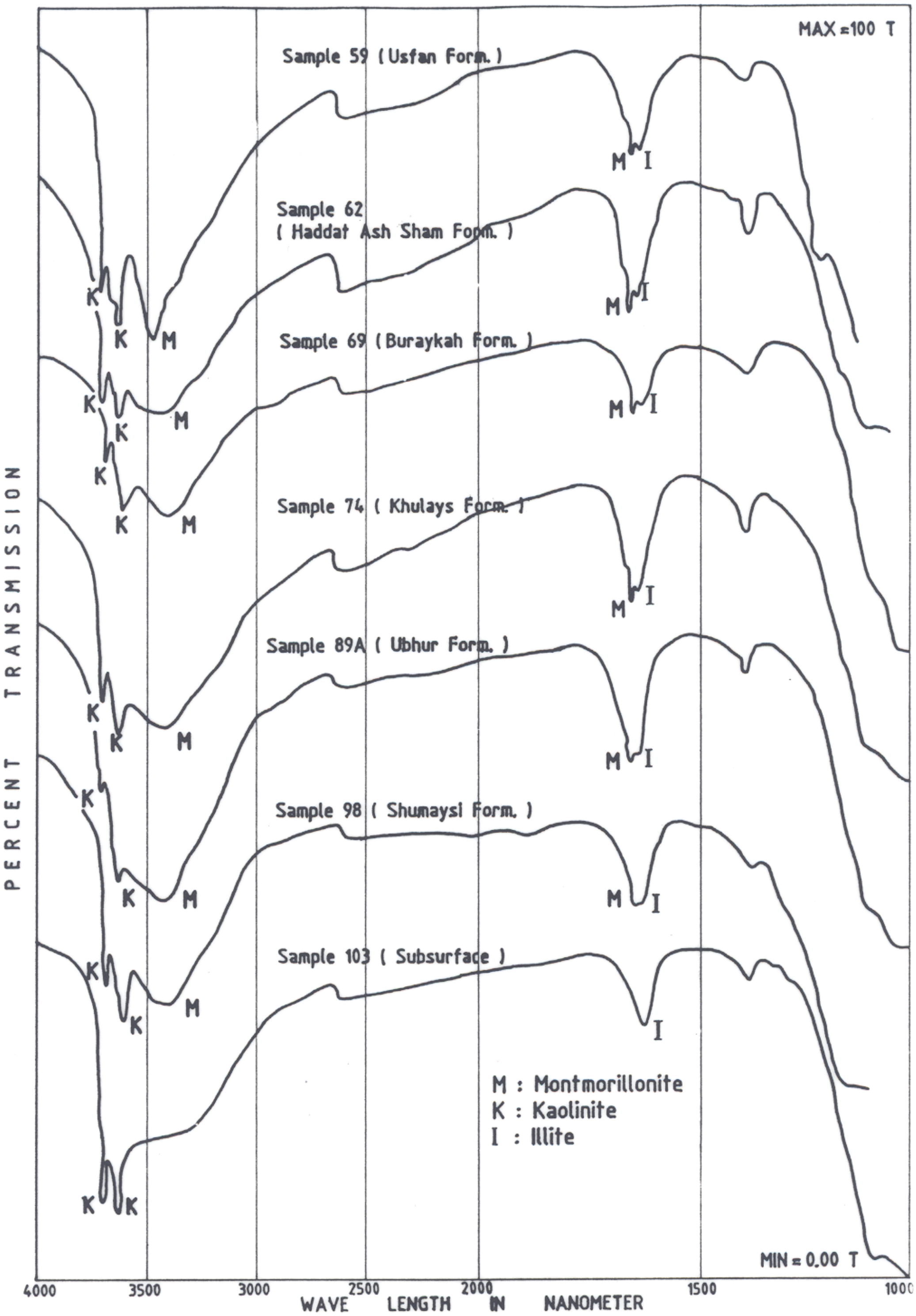

Figure 7. Infrared spectra of representative clay samples.

7. Conclusions

1- Results arrived at by the various mineralogical techniques agree with each other concerning the composition of the investigated Tertiary clay deposits.Generally these deposits are predominantly composed of montmorillonite Ca ++ and/or Mg ++ rich variety) and kaolinite with subordinate illite and minor chlorite. Mixed layer montmorillonite-illite is recorded in two samples, one belongs to Haddat Ash Sham Formation and the other is a subsurface sample .

2- Based on the relative abundance of the identified minerals by X-ray diffraction analysis, the studied clay deposits can be classified into three varieties. The first, which is the most common, is highly montmorillonitic (e.g., most of the clay samples of Khulays and Buraykah formations and some of the Usfan Formation). The second is made up of a mixture of montmorillonites followed by kaolinite and illite (e.g., most of the clay samples of Haddat Ash Sham and Daffin formations and some of the Usfan Formation). The last is highly kaolinitic and sometimes with lesser montmorillonite (e.g., clay deposits of Shumaysi Formation and some of those from subsurface).

3- It is observed that kaolinite content increases southward where it represents the only component of some of the Shumaysi Formation samples in Makkah quadrangle. On the other hand, chlorite increases, as a minor component, northward where it is recorded in most samples of the Buraykah and Daffin formations in Rabigh quadrangle.

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Received 27 February 2002
Accepted 1 November 2002