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PALEOZOIC CLAYS OF TUNISIA

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Abstract: The oil drilling holes carried out on the Saharan Platform (southern Tunisia) produced evidence of the presence of Paleozoic bedrock. The dominant lithology of the Lower Paleozoic is clays and sandstones whereas the Upper Paleozoic is marked by carbonated sandstone and clayey marine series. The qualitative and semiquantitative mineralogical compositions (bulk rock and clay fraction) were evaluated by X-ray diffraction, using Siemens Kristalloflex 810 diffractometer. The chemical analyses of major elements were carried out by Atomic Absorption Spectrophotometry. Trace and rare earth elements were determined by ICP-MS Perkin-Elmer SCIEX Elan-5000. Chemical microcomposition was determined using Phillips CM20 STEM. The mineralogical studies of Paleozoic clays showed the predominance of illite accompanied by small amounts of kaolinite and chlorite. The geochemical data put in evidence the relatively high amounts of Fe_2O_3 and Na_2O which are most probably associated with aluminosilicates. The flat pace of the rare earth curve analyzed in relation to the NASC indicates the tendency to the impoverishment in HREE.

Key words: Paleozoic, Tunisia, geochemistry, mineralogy, clays.

Introduction

The studied region is located close to the southern border of Tunisia corresponding to the Saharan Platform domain (Fig. 1).

The Precambrian bedrock is covered by Paleozoic sub-tabular clay and sand series, which have been sealed unconformably by a Mesozoic succession of strata (Berkaloff 1933; Bonefous 1963; Busson 1967; Memmi et al. 1986; Bouaziz 1995). The Paleozoic series related to the oil drilling in the south of Tunisia were studied from the mineralogical and geochemical point of view.

Geological setting

Precambrian and Paleozoic formations do not outcrop on the surface in Tunisia. The only exception is the Permian deposit outcropping close to Djebel Tebaga, west of Medenine. However, the oil drilling works carried out in the South of Tunisia crossed the different periods of the Paleozoic (Memmi et al. 1986; Ben Ferjani et al. 1990).

The Cambrian is represented by a thick set of sandstone assigned to the Sidi Toui Formation. This unit rests in discordance on the crystalline and metamorphic bedrock (Memmi et al. 1986).

The series assigned to the Ordovician consist essentially of clay and sandstone in the basis, evolving to a clay-conglomeratic series in the top (Memmi et al. 1986; Jamoussi 2001).

The Silurian starts with an essentially clayey sequence, radioactive and rich in organic matter. It has been assigned to the Tannezouft Formation. The succession becomes enriched itself in sandstone components forming the Acacus Formation (Jeager et al. 1995).

The Devonian is found in the south of the Dahar arc. The Devonian marks the return to deposits of clay and sandstone.

The Carboniferous brought a return to more straightforwardly marine conditions. It is found in the Southern border of Tunisia and NE of the Saharan Platform, represented by clay, limestone and sandstone.

The Permian occurs only in the North of the Saharan Platform, in the furrow of the Jeffara where it can reach some important thickness. The Permian of the Tebaga Formation, of which only the top section outcrops in the Northwest of Medenine, constitutes the only African outcrop of marine Permian represented by bioclastic limestone, argillaceous limestone and clay (Khessibi 1985).

Permo-Triassic deposits of the Haïrech, located in the Northern Atlas, were studied for comparison. They are formed by a thick formation of slightly metamorphized, pelitic sandstone. It is a regular sequence of thin sandstone and clay in thin beds (Alouani & Tlig 1988).

Materials and methods

Several samples of drill cores BMT1 and LG2 (see Fig. 1 for location), were studied as representatives of the different Paleozoic formations.

The qualitative and semiquantitative mineralogical compositions (bulk rock and clay fraction) were evaluated by X-ray

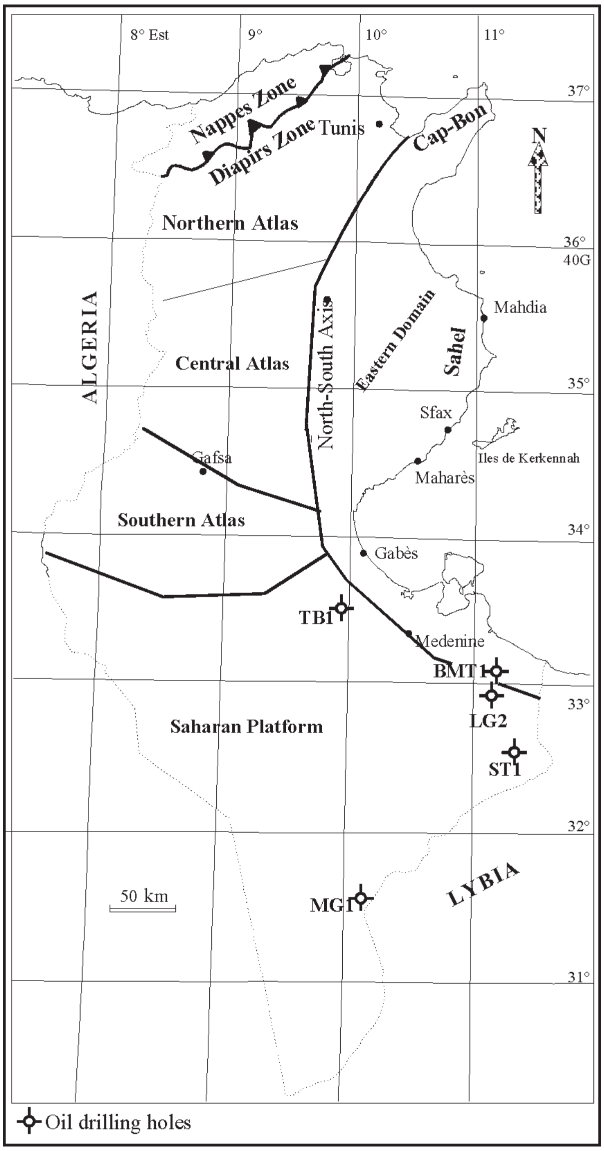

Fig. 1. Location of the studied zone.

diffraction, using a Siemens Kristalloflex 810 diffractometer, Cu-Kα radiation, by the powder method (bulk rock) and as oriented preparations (clay fraction), air-dried, saturated with ethylene glycol and heated to 550 °C.

Quantification of different phases was carried out by the classic method of diffraction peak area measurement and reflection power (Millot 1964; Schultz 1964; Biscaye 1965; Barahona 1974; Mellinger 1979; Brindley & Brown 1980; Caillere et al. 1982; Pevear & Mumpton 1989).

The chemical analyses of major elements were carried out by Atomic Absorption Spectrophotometry. Trace and rare earth elements were determined by a ICP-MS Perkin-Elmer SCIEX Elan-5000, using Rh and Re as internal standards. Accuracy was 2% and 5% for 50 ppm and 5 ppm, respectively. The detection limits of elements were 100 ppb for REE and Th, 5 ppm for transition elements and Cs, Rb, Sr, Ba and Pb, and 10 ppm for Li. Chemical microcomposition was determined using a Phillips CM20 STEM.

Results and discussion

The mineralogical studies of Paleozoic clays showed the general predominance of siliciclastic minerals in the bulk-rock compositions and of illite in their clay fractions (Fig. 2 and Table 1).

Analysis of the mean values of the bulk-rock mineralogical composition for each one of the stratigraphic units revealed an almost regular alternation of phyllosilicate rich units with units rich in quartz. Only the Lower Permian and Upper Carboniferous are enriched in carbonate minerals (mainly dolomite).

The clay fractions show (Fig. 2 and Table 1) the general predominance of illite associated with kaolinite and chlorite. Upper Carboniferous, Lower and Upper Silurian formations are the only exceptions to this rule.

Clay mineral association (illite + kaolinite + chlorite) characterizes the Cambrian-Ordovician and Lower Ordovician pe-

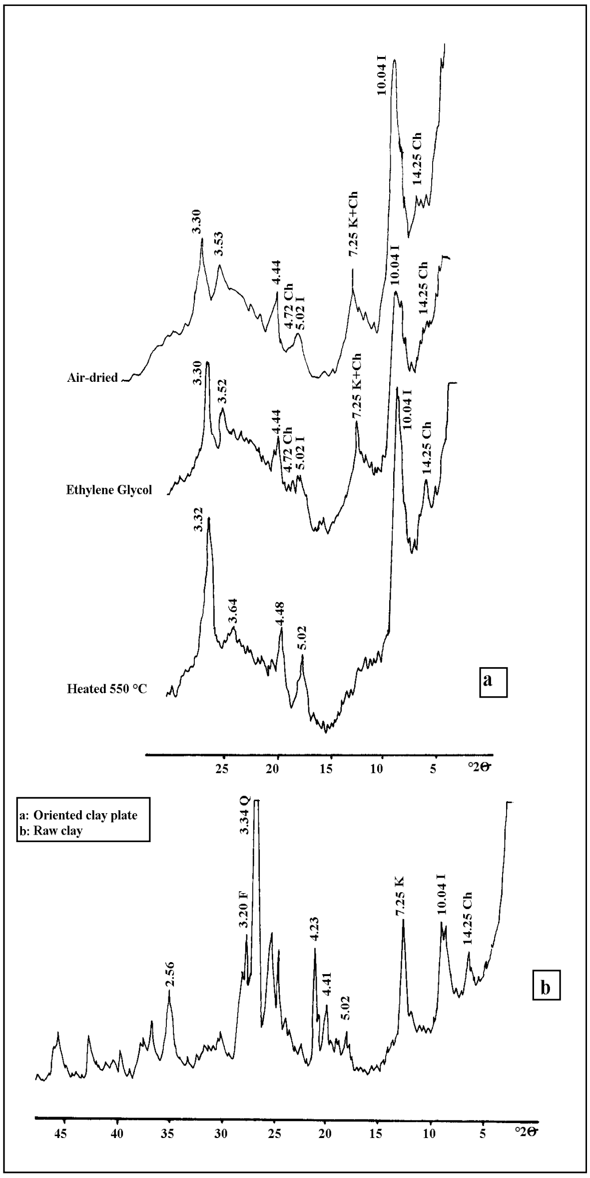

Fig. 2. XRD patterns of Carboniferous clay.

Table 1: Mineralogical composition of the studied Paleozoic sequences of the Saharan Platform. (Phyll — phyllosilicate, Ca — calcite, Dol — dolomite, Qz — quartz, Gyp — gypsum, Feld — feldspar, Sm — smectite, Ill — illite, Kaol — kaolinite, Ill/Sm — interstratified illite smectite, Chl — chlorite.)

Stratigraphic unit	Bulk-rock Mineralogy						Clay-fraction Mineralogy				
	Phyll	Ca	Dol	Qz	Gyp	Feld	Sm	Ill	Kaol	Ill/Sm	Chl
Permo-Triassic - Hirech	57	0	0	36	0	7	0	97	3	0	0
Permian - Dkilet Toujan	81	5	0	8	5	2	11	45	18	21	6
Upper Permian	59	4	8	26	0	3	0	75	14	0	11
Lower Permian	20	0	74	5	0	1	0	83	17	0	0
Upper Carboniferous	38	0	30	31	0	1	0	36	55	0	9
Lower Carboniferous	50	0	0	45	0	5	0	75	14	0	11
Upper Devonian	55	0	0	38	0	7	0	77	13	0	10
Lower Devonian	45	1	1	52	0	1	0	80	12	0	8
Upper Silurian	51	1	0	44	0	4	0	37	45	0	18
Lower Silurian	58	1	0	39	0	2	0	42	49	0	9
Lower Ordovician	23	1	1	70	0	5	0	80	12	0	8
Cambrian-Ordovician	34	1	1	46	0	18	0	77	13	0	10

riod. Chlorite is of Fe-type and, according to Giblin & Cahen (1982) and Jamoussi (2001), essentially authigenic. These detrital siliciclastic deposits evolve from finer ones at the base to coarser ones (conglomeratic) towards the top. This sedimentological evolution is also recorded in the mineralogical composition by the enrichment in quartz (bulk-rock) and in illite (clay fraction), as well as by the impoverishment in feldspars and chlorites. These deposits have been considered to be periglacial marine sediments, low energetic at the base and high energetic at the top.

Silurian (Upper and Lower) deposits are finer and richer in phyllosilicates, presenting the enrichment in kaolinite, associated with illite and chlorite, the latter being essentially authigenic, increasing from the Lower Silurian to the Upper Silurian. This mineralogical signature points out to environmental changes occurring in the Lower Silurian, more favourable to chemical alteration and transformation, vanishing during the Upper Silurian.

Clays of the Devonian show again a clear predominance of illite with little kaolinite and chlorite (Jamoussi 2001), the deposits being essentially the product of fresh detrital continental supplies.

From the Lower Carboniferous to the Upper Carboniferous the strong increase in dolomite (bulk-rock) and in kaolinite (clay fraction) contents is counterbalanced by the decrease in quartz, illite and chlorite contents. These carbonate deposits are clearly marine, brackish, deposited in relatively confined waters.

The Permian shows again a predominance of illite accompanied by kaolinite and also, in Upper Permian, by chlorite; in some Permian outcrops smectite and illite-smectite are present, decreasing then the relative illite and chlorite contents (Table 1). These deposits are mainly continental with lagoonal episodes, presenting some marine domains (carbonate platforms, reefs), the sediments having not been submitted to substantial pressure and temperature increases.

The chemical analyses of the major elements (Table 2) were performed only in the bulk-rock samples. It can be noticed that Al_2O_3 presents a strong interrelationship with Fe_2O_3 (r = 0.64 correlation factor) and Na_2O (r = 0.62), which confirms that these elements are contained in aluminosilicate minerals. On the other hand, Al_2O_3 content presents a very weak interrelationship with K_2O (r = 0.03).

Trace element contents (Tables 3a and 3b; Fig. 3) show a regular behaviour for five analyzed stratigraphic intervals: Cambrian-Ordovician, Lower Devonian, Upper Permian, Permian and Permo-Triassic, the latter showing a slight impoverishment in relation to the others (see SP5 at Fig. 3).

Rare earth element (REE) contents (Table 4) are plotted after NASC normalization (Fig. 4), showing a practically flat

Table 2: Chemical composition (major elements in %) of the studied Paleozoic sequences of the Saharan Platform.

Stratigraphic unit	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	$SO^{=}_3$	IL	SiO_2/Al_2O_3	Fe_2O_3/Al_2O_3	Al_2O_3/K_2O
Permo-Triassic - Hirech	72.91	12.68	4.14	0.18	0.34	0.54	4.22		2.76	5.75	0.33	3.00
Permian - Dkilet Toujan	42.50	17.44	8.65	6.44	2.28	0.50	3.86	3.56	14.42	2.44	0.50	4.52
Upper Permian	47.14	16.58	5.92	4.93	3.53	1.17	3.60	2.51	13.34	2.84	0.36	4.61
Lower Permian	15.40	7.90	3.43	20.16	14.59	0.15	1.88	0.00	34.76	1.95	0.43	4.20
Upper Carboniferous	46.29	16.98	5.57	7.14	4.97	0.16	3.69	3.59	10.49	2.73	0.33	4.60
Lower Carboniferous	49.42	20.04	6.15	6.31	2.58	1.40	5.78	1.65	6.48	2.47	0.31	3.47
Upper Devonian	50.93	22.67	7.72	0.35	1.58	1.19	2.58	0.65	11.69	2.25	0.34	8.79
Lower Devonian	55.34	20.78	6.00	2.25	1.11	0.84	2.77	1.06	9.15	2.66	0.29	7.50
Upper Silurian	50.85	20.42	11.44	0.71	1.63	1.46	2.53	0.74	10.18	2.49	0.56	8.07
Lower Silurian	50.99	21.73	9.58	0.50	1.14	1.79	2.41	0.79	10.48	2.35	0.44	9.02
Lower Ordovician	57.13	17.85	5.38	3.29	1.54	0.62	4.82	1.21	8.05	3.20	0.30	3.70
Cambrian-Ordovician	61.98	18.72	5.58	0.17	1.28	0.88	5.78	0.00	3.26	3.31	0.30	3.24

Table 3a,b: Chemical composition (trace elements in ppm) of the studied Paleozoic sequences of the Saharan Platform.

a

Stratigraphic unit	Sample	Li	Rb	Cs	Be	Sr	Ba	Sc	V	Cr	Co	Ni	Cu	Zn
Permo-Triassic Hirech	SP5	5.3	108.2	3.7	2.2	13.8	253.1	12.1	91.5	150.4	1.4	34.9	2.3	1.7
Permian Dk. Toujane	Pr1	54.0	151.1	8.1	3.1	272.0	190.1	22.2	164.8	149.6	10.7	66.4	13.6	65.1
Upper Permian	PRM3	118.2	217.2	17.9	2.6	167.4	191.3	18.6	200.5	148.8	6.4	42.8	14.5	48.7
Lower Devonian	DV2	111.0	129.1	10.9	2.9	65.4	137.9	20.2	151.1	129.5	12.0	54.9	26.6	65.2
Cambrian-Ordovician	ORD1	34.5	330.0	19.4	3.6	34.5	316.0	17.3	114.6	134.6	5.8	24.0	7.6	43.8

b

Stratigraphic unit	Sample	Ga	Y	Nb	Ta	Zr	Hf	Mo	Sn	Tl	Pb	U	Th
Permo-Triassic Hirech	SP5	17.7	4.2	2.6	0.2	2.2	0.1	0.5	4.1	0.3	2.4	0.7	7.0
Permian Dk. Toujane	Pr1	28.4	18.2	13.8	1.0	103.9	3.1	2.0	7.2	0.7	18.4	2.4	14.4
Upper Permian	PRM3	35.9	10.5	9.8	0.7	89.9	2.8	2.2	94.2	1.0	12.5	1.9	10.1
Lower Devonian	DV2	35.5	15.4	10.5	1.3	51.5	1.7	2.0	39.9	0.8	27.8	1.5	17.8
Cambrian-Ordovician	ORD1	35.2	10.1	12.9	1.5	34.9	1.4	2.0	80.5	1.4	11.2	4.0	19.8

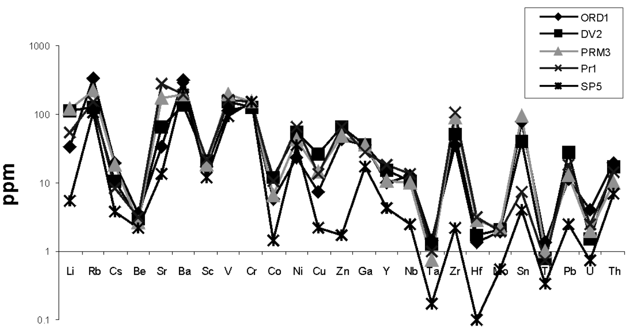

Fig. 3. Trace element distribution curves.

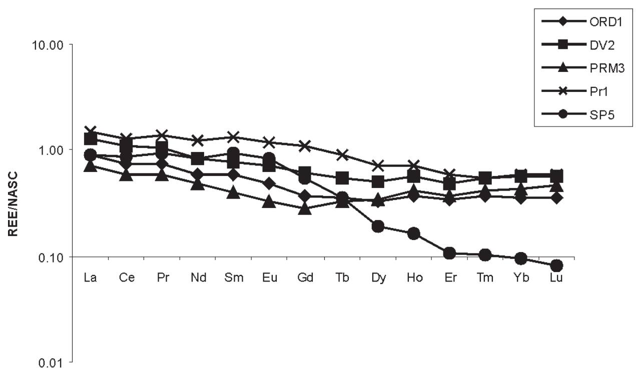

Fig. 4. Rare earth element distribution curves (NASC — normalized).

Table 4: Chemical composition (rare earth elements in ppm) of the studied Paleozoic sequences of the Saharan Platform.

Sample	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
SP5	27.55	62.48	7.31	27.97	5.26	1.03	2.83	0.31	1.06	0.17	0.36	0.05	0.30	0.04
Pr1	45.45	93.38	10.92	40.80	7.55	1.47	5.63	0.78	3.98	0.74	2.00	0.28	1.86	0.28
PRM3	21.36	42.34	4.61	15.83	2.27	0.41	1.49	0.28	1.86	0.43	1.26	0.21	1.34	0.23
DV2	38.55	79.76	8.17	27.10	4.45	0.89	3.15	0.47	2.76	0.59	1.66	0.27	1.75	0.27
ORD1	26.76	55.24	5.93	19.37	3.38	0.61	1.94	0.31	1.83	0.39	1.15	0.18	1.10	0.17

tendency of lines, but putting in evidence of some impoverishment in heavy rare earth elements (HREE), in particular for the Permo-Triassic (SP5).

Table 5 shows the structural formulas of clay minerals from some of different studied stratigraphic formations. These structural formulas were computed supposing that all chemical charges are compensated. For illite, iron is regarded as trivalent, for chlorites as divalent.

Structural formula of one of the Ordovician illites:
$K_{0.75}\,Mg_{0.05}\,(Al_{1.73}\,Mg_{0.11}\,Fe^{3+}{}_{0.20})\,(Si_{3.16}\,Al_{0.84})\,O_{10}\,OH_2$

Structural formula of one of the Silurian illites:
$K_{0.82}\,(Al_{1.63}\,Mg_{0.23}\,Fe^{3+}{}_{0.21})\,(Si_{3.21}\,Al_{0.79})\,O_{10}\,OH_2$

Structural formula of one of the Silurian paragonites:
$K_{0.35}\,Na_{0.18}\,Mg_{0.09}\,(Al_{1.81}\,Fe^{3+}{}_{0.29})\,(Si_{2.99}\,Al_{1.01})\,O_{10}\,OH_2$

Structural formula of one of the Permian chlorites:
$K_{0.55}\,(Al_{1.51}\,Mg_{0.18}\,Fe^{2+}{}_{2.25}\,Ti_{0.05})\,(Si_{3.60}\,Al_{0.40})\,O_{10}\,OH_8$

Structural formula of one of the Permo-Triassic illites:
$K_{0.72}\,Mg_{0.01}\,(Al_{1.53}\,Mg_{0.37}\,Fe^{3+}{}_{0.18}\,Ti_{0.01})\,(Si_{3.30}\,Al_{0.70})\,O_{10}\,OH_2$

The studied samples of the Tunisian Paleozoic present illites showing irregular degrees of crystallinity, as can be seen from Figure 5. The majority of the analyzed samples shows Kubler Crystallinity Index for illite corresponding to medium up to well ordered values which are located in the zone of diagene-

Table 5: Numbers of ions in crystal structure formulas of clay minerals from the studied stratigraphic formations.

Samples	Tetrahedral sheet		Octahedral sheet				Interlayer cations			
	Si	Al IV	Al VI	Mg	Fe	Ti	K	Ca	Na	Mg
Ord1/1 illite	3.11	0.89	1.61	0.18	0.27	0.00	0.87	0.00	0.00	0.01
Ord1/2 illite	3.21	0.79	1.85	0.04	0.12	0.00	0.64	0.00	0.00	0.08
Mean ORD1 illite	3.16	0.84	1.73	0.11	0.20	0.00	0.75	0.00	0.00	0.05
SLR2/2 illite	3.21	0.79	1.63	0.23	0.21	0.00	0.82	0.00	0.00	0.00
SLR2/3 illite-paragonite	2.99	1.01	1.81	0.00	0.29	0.00	0.35	0.00	0.18	0.09
Pr1/2 chlorite	3.60	0.40	1.51	0.18	2.25	0.05	0.55	0.00	0.00	0.00
SP5/1 illite	3.27	0.73	1.46	0.45	0.21	0.01	0.76	0.00	0.00	0.00
SP5/2 illite	3.38	0.62	1.50	0.44	0.16	0.01	0.72	0.00	0.00	0.00
SP5/3 illite	3.25	0.75	1.64	0.23	0.19	0.02	0.67	0.00	0.00	0.04
Mean SP5 illite	3.30	0.70	1.53	0.37	0.18	0.01	0.72	0.00	0.00	0.01

Table 6: Non clay minerals and clay minerals assemblages of the Tunisian Paleozoic clayey units, and paleoambiental interpretations. (Phyll — phyllosilicate, Ca — calcite, Dol — dolomite, Qz — quartz, Gyp — gypsum, Feld — feldspar, Sm — smectite, Ill — illite, Kaol — kaolinite, Ill/Sm — interstratified illite smectite, Chl — chlorite).

Stratigraphic units	Bulk-rock phases	Clay-fraction	Paleoambiental interpretations
Permian- Dkilet Toujan	Phyll, Qz, Ca	Ill, Sm/Ill-Sm, Kaol	Continental, lagoonal episodes
Upper Permian	Phyll, Qz, Dol	Ill, Kaol, Chl	Marine, deep sea
Lower Permian	Dol, Phyll	Ill, Kaol	Carbonated platforms, reef
Upper Carboniferous	Phyll, Qz, Dol	Kaol, Ill, Chl	Marine, brackish, confined
Lower Carboniferous	Phyll, Qz, Feld	Ill, Kaol, Chl	Transition environment
Upper Devonian	Phyll, Qz, Feld	Ill, Kaol, Chl	Decrease of the detrital supply
Lower Devonian	Qz, Phyll	Ill, Kaol, Chl	Fresh detrital continental supply
Upper Silurian	Phyll, Qz	Kaol, Ill, Chl	Vanishing of chemical alteration
Lower Silurian	Phyll, Qz	Kaol, Ill, (Chl)	Increase of chemical alteration
Lower Ordovician	Qz, Phyll, (Feld)	Ill, Kaol, Chl	Periglacial marine, high energetic
Cambrian-Ordovician	Qz, Phyll, Feld	Ill, Kaol, Chl	Periglacial marine, low energetic

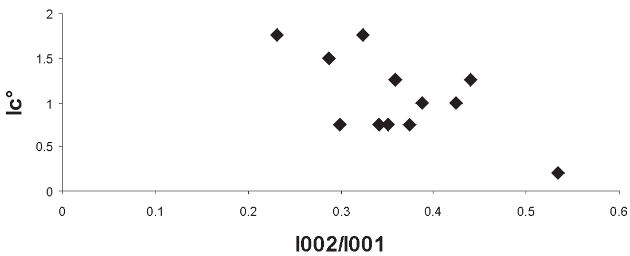

Fig. 5. Illite crystallinity (Kubler index vs. Esquevin index).

sis but coming closer to the anchizone (Fig. 5). The only sample plotted clearly in the epizone belongs to the Permo-Triassic of Hirech confirming the sudden metamorphism presented by these levels.

A synthesis of the mineralogical characterization of Tunisian Paleozoic clayey units is shown in Table 6, as well as the proposal of some paleoenvironmental inerpretations.

Conclusions

Paleozoic clays from Tunisia sampled in oil drill cores were studied. The mineralogy of these clays shows a predominance of well crystallized illite accompanied by smaller contents of kaolinite and chlorite. Fe_2O_3 and Na_2O contents are essentially concentrated in aluminosilicates. The rare earth element representation in relation to the NASC, shows the practically flat curves and the tendency to the impoverishment in heavy rare earth elements.

Clayey Paleozoic sediments are produced from the alteration of the crystalline basement and subsequently influenced by diagenetic transformation. The higher illite crystallinity shown in certain strata indicates on the one hand the diagenesis action and on the other hand the influence of diverse orogeneses rejuvenated the relief consequently favouring the transport of detrital materials. Clay mineral assemblages reflect the climatic oscillations as well as the effects of tectonic events.

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