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77 - 84

STUDY OF SOME SMECTITES DERIVED BY WEATHERING OF CRYSTALLINE ROCKS OF THE PROVENCE REGION (SE FRANCE) INFLUENCE OF THE PARENT ROCK ON THEIR CHEMICAL COMPOSITION

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Abstract: The saprolites from igneous rocks of the Provence basement (SE France) are poor in $< 2\ \mu$m fraction (1 - 5 %). Clay minerals consist of smectites, illite, kaolinite, metahalloysite, some vermiculite and an appreciable proportion of interstratified clays (20 - 30 %). In this paper, we will study especially the smectites which are composed of two groups: the first one derived from granite, diorite and basalt, is a montmorillonite type, without tetrahedral substitution and whose sum of octahedral cations varies from 2.0 to 2.2. The second group, derived from trachydolerite, is intermediate with tetrahedral substitution, like in beidellites. The cationic octahedral content is 2.5. The chemical composition of both groups shows a clear influence of the parent rock. The different smectites of this area were influenced by the ferromagnesian and aluminomagnesian minerals present in the parent rocks.

Key words: weathering, igneous rocks, Provence (SE France), dioctahedral and intermediate smectites.

Introduction

The geological study of the Provence basement in the south France, has been conducted by several authors: Boucarut (1971), Pupin & Turco (1973), Pupin (1976). The scientists have shown the variety of the composition of these igneous rocks composed of very diversified minerals, from acid to basic types, of volcanic to plutonic origins and variable age from 225 to 340 Ma. The rocks, submitted for a long time to weathering, have undergone a more or less deep alteration according to the situations, giving rise to saprolites poor in fine fraction $< 2\ \mu$m - Chevalier (1984). This author has shown the particular complexity of the mineralogical composition of this fraction $< 2\ \mu$m derived from the weathering of primary minerals. The clays identified are in effect: smectites, illites, kaolinites, metahalloysites, some vermiculite and an appreciable proportion (20 - 35 %) of mixed layer clays (10 - 14 V, 10 - 14 M, 14 C - 14 V, 14 C - 14 M) in which the vermiculite type predominates in acid rocks and the smectite type in basic rocks. Sometimes, smectite is almost the exclusive compound of the fraction $<2\ \mu$m, for example from the trachydolerites, or they are associated with illites.

In this paper, we will only study the composition of the smectites and their connection with the parent rocks, prolonging in this way our own researchs (Chevalier et al. 1982; Chevalier 1984).

Geographical localization of the study

The 2/1 minerals studied have been extracted from the fraction of saprolites developed from four different parent rocks.

a - hololeucratic alkaline granite;
b - quartzitic diorite;
c - quartzitic basalt, close to trachyandesite;
d - trachydolerite.

Fig. 1 indicates the main geological facies of the Provence and the localization of the studied profiles, while the Tab. 1 gives the ages of the four rocks, estimated by K-Ar method.

Mineralogical and chemical composition of the parent rocks

The Moulin Blanc Granite

Occupying the northern area of the Saint-Tropez peninsula, the Moulin Blanc Granite is an alkaline holo-

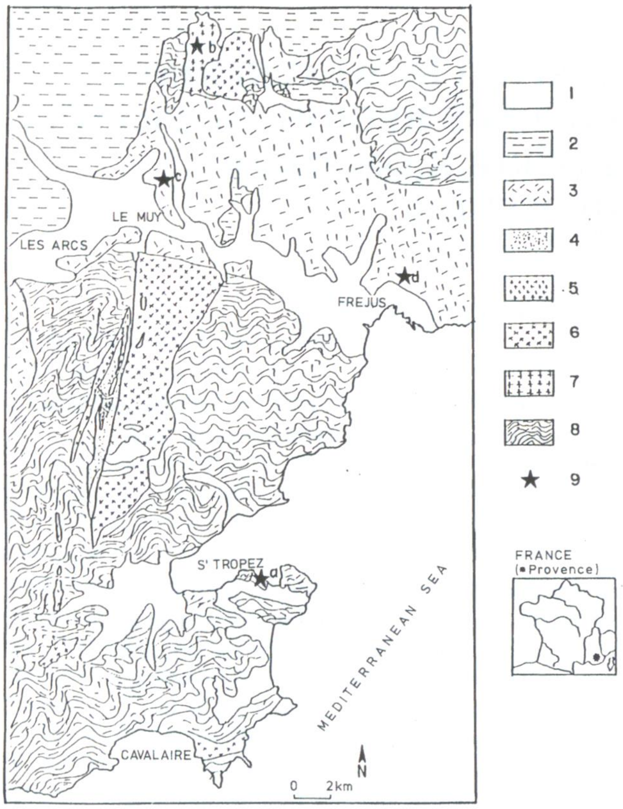

Fig. 1. Simplified geology and map of Maures, Tanneron and Esterel Massif (Provence, SE France). **1** - Quaternary; **2** - Post-Permian rocks; **3** - Permian volcanoclastic rocks; **4** - Carboniferous rocks; **5** - Plan de la Tour microgranite; **6** - Granite; **7** - Gabbro and quartz diorites; **8** - Metamorphic rocks; **9** - Samples: **a** - Moulin Blanc Granite; **b** - Prignonet Diorite; **c** - Col du Mistral Basalt; **d** - Lieutenante Trachydolerite.

leucratic rock, with a medium grained structure. The plagioclase is an oligoclase An_{14} and is rich in muscovite. It contains a very small fraction of cordierite (0.4 %), which manifests a richness of the rock into Al_2O_3, traces of andalusite and also sillimanite (Pupin 1976). Traces of hydrothermal calcite are observed inside oligoclase.

The Prignonet Diorite

It crops out on three km in breath from west to east in the Tanneron Massif. It is a dark rock, with an equigranular or planar structure, admitting a wide variety of facies, from granodiorites to quarzitic diorites and gabbros. The material selected for this study is a tonalite rich in quartz (20 - 25 %). Under microscope, the crystals seem broken into little fragments more or less recrystallized.

The plagioclase is an andesine An 41 - 48; the biotite is very well represented (10 - 20 %) while the green hornblende varies between 3 and 10 % in the massif. The K-feldspars are not higher than 5 % of the total of the rock.

The "Col du Mistral" Basalt

The rock is extracted from a lawa flow of 15 km in length in the Esterel Massif between the Ajay (SE) and the Frejus (W) valleys. Its thickness ranges from 20 to 50 meters. The texture oscillates between microlitic and doleritic in the center of the lava flow and fluidal microlitic at the edges. This material is, in point of fact, close to trachyandesites, according to the mineralogical parameters.

The plagioclase is an andesine An_{36}. The rock is poor in CaO, and very rich in TiO_2, showing the abundance of the ilmenite. Quartz is always present. The mesostase, weakly represented, is composed especially of chlorite and traces of sanidine.

The "La Lieutenante" Trachydolerite

Situated in the Esterel Massif, North of Frejus, the trachydolerite is an interstratified seam inside red pelites of the end of Lower Permian.

The plagioclases (andesine $An_{34\text{-}37}$), are the fundamental minerals of the trachydolerite, with their appearance of laths with a variable size. The olivine is abundant, often serpentinised. The pyroxenes are always absent. The opaque minerals are represented especially by ilmenite whose presence is confirmed through the TiO_2 richness of the rock. Chlorite, apatite and secondary calcite are also found.

The mineralogical and chemical composition of these four rocks are summarized in Tabs. 2 and 3, with the different and characteristic parameters.

Analytical methods

The fine fraction $< 2\ \mu m$, separated from the saprolites by sedimentation, has been studied by the classi-

Table 1: Age of geological studied formations.

ROCK	LOCALISATION	AGE(1)(Ma)	GEOLOGICAL PERIOD	REFERENCES
a) Granite	Moulin Blanc	>305 (2)	Namurian-Westphalian	Chevalier (1984)
b) Quartzitic diorite	Prignonet	340–380	Upper Devonian Dinantian	Chevallier (1984)
c) Basalt (lava-flow)	Col du Mistral	241 ± 10	Low to Middle Trias	Roubault et al. (1970)
d) Trachydolerite (Seam)	La Lieutenante	228–241	Low to Middle Trias	Boucarut (1971)

(1) Ma = Million Years

(2) No date, but Moulin Blanc Granite is older than Camarat Granite, adjacent in the Saint-Tropez Peninsula, dated of 305 Ma.

Table 2: Mineralogical composition of parent rocks (%).

SAMPLE	a) GRANITE Moulin Blanc	b) DIORITE Prignonet	c) BASALT (1) Col du Mistral	d) TRACHYDOLERITE (2) La Lieutenante
Major elements				
Quartz	33.60	20.00	7.00	
K-Feldspars	23.00	4.60	7.00	
Plagioclases Oligoclase	27.70			
Andesine		48.20	35.20	56.00
Biotite	2.00	19.20		
Chlorite			16.30	6.40
Muscovite	10.00			
Hornblende		3.30		
Olivine			20.00	15.50
Opaque minerals (especially ilmenite)	0.10	1.30	14.00	12.50
Minor elements				
Cordierite	0.20			
Andalusite	0.20			
Apatite	0.40	0.40	0.80	0.90
Secondary Calcite	0.10			
Remaining Al_2O_3		1.10		
Free Feoxide	0.20	2.70		
TOTAL	97.50	100.80	100.30	95.50

(1) The rock is, in fact, a trachyandesite.
(2) Weakly weathered rock.
(3) Results extracted from the thesis of Chevalier (1984).

cal methods described in the previous works (Chevalier 1984): X-rays diffraction, the peak (060) being used in order to identify the dioctahedral or trioctahedral character of the smectites, test Li (Greene-Kelly 1953; Hoffman-Klemen 1960), thermal analysis, IR spectroscopy.

In this especially physical and chemical study of the smectites, the chemical analysis is carried out by atomic absorption spectroscopy after attack of the sample with nitric and hydrofluoric acids, dissolution in HCl and buffered with $SrCl_2$. From this total analysis, the chemical composition of the smectites is obtained by elimination of the goethite identified by differential thermal analysis through the measurement of free iron Fe_2O_3 (Deb 1950) which is a reduction of iron by the citrate-dithionite. The kaolinite is estimated from the loss of water between 400 and 600 °C of the thermogravimetric curve and the illite from the K_2O content: 1 % K_2O corresponding about to 10 % illite (Mehra-Jackson 1959).

It is clear that the result obtained and subsequently the structural formula of smectites with 11 oxygens for basis, is only approximate, because the free iron is not included in totality in the goethite and otherwise, the whole K_2O does not enter in the constitution of the illite, one fraction, probably very weak, constituting the structure of the smectite.

The parameters *a* and *b* of the smectites (in Å) have been estimated from their chemical composition (Brown 1961). It leads to the evaluation of the total surfaces of clays from the formula:

$S\ m^2/g = a \times b \times N/\ \text{molecular weight} \times 10^{20}$

$N = \text{Avogadro constant} = 6.02 \times 10^{23}$

At least, the cation exchange capacity (CEC) has been determined approximately from the charge of layer: CEC = 200 × charge (me/100 g)(Foster 1951).

Results

They are represented in the Tabs. 4 - 6, with the mineralogical composition of the fraction $< 2\ \mu m$ of different profiles, the structural formula of smectites, the charge in the tetrahedral, octahedral and interlayer positions, and some physical and chemical characteristics of the 2/1 clays.

A continual increase of the proportion of smectite is observed with the basicity of the parent rock which becomes preponderant in the saprolites derived from the basalt and the trachydolerite, the goethite follows a similar evolution. The illite, widely dominant in the granite (74 %) represents only 1/4 of the fraction $< 2\ \mu m$ in the trachydolerite and becomes very low in the basalt. Relating to kaolinite it is represented especially in the diorite and the basalt.

The X-rays allow to identify all 2/1 minerals as smectites: 14 Å and swelling with glycerol treatment. But the

Table 3: Chemical composition of parent rocks.

OXIDES %	a) GRANITE Moulin Blanc	b) DIORITE Prignonet	c) BASALT Col du Mistral	d) TRACHYDOLERITE La Lieutenante
SiO_2	71.75	58.15	51.45	44.80
Al_2O_3	14.42	16.80	15.63	16.40
Fe_2O_3	-	3.18	0.26	2.98
FeO	0.78	3.75	6.58	4.77
CaO	0.97	5.60	1.70	2.29
MgO	0.23	4.18	6.89	8.80
K_2O	4.80	2.40	1.16	0.76
Na_2O	2.83	3.12	3.27	5.87
TiO_2	0.09	0.61	5.29	4.82
P_2O_5	0.20	0.20	0.36	0.45
H_2O^+	1.02	1.06	4.82	6.49
H_2O^-	n.d	n.d	1.89	0.73
TOTAL	97.09	99.05	99.30	99.17
Parameters CPIW				
p	I	II	II'	II'
q	3 (4)	4 (5)	4	5
r	1 (2)	3	2	2
s	3	'4	4	'5
h	1'	2	2'	3
k	1	1	1	1
l	1	1	1	1
m	3 (4)	2'	2 (3)	2
Al/Alc	1.46	2.17	2.35	1.56
Al/Calc	1.23	0.93	1.60	1.11
Qz = Si - Si' (free silice)	197.2	24.1	15.5	-47.6
Parameters JUNG-BROUSSE				
SAT	38	15.6	24	0
COL	9.5	22.7	45.5	47.3
FELDS	50.7	21.7	16	7.8

Results extracted from the thesis of Chevalier (1984), p. 30, 41 and 45.

Li-test differentiates clearly two groups of smectites: the first one formed by the clays derived from granite, diorite and basalt gives by X-rays a fundamental peak at 17.65 Å, which after 24 hours heating at 300 °C is crushed to 10 Å. This characteristic shows the occurrence of montmorillonites. On the contrary, for the Lieutenante smectites, derived from the trachydolerite, the 17.6 Å peak is maintained after heating. So, these clays are close to beidellites.

The charge of layer (Tab. 5) corresponds perfectly to that of smectites for all minerals, being always included between 0.2 and 0.6 (Mering-Pedro 1969). But the structural formulae (Tab. 5) also differentiate the two groups of smectites: the first one is dioctahedral with a sum of cations included between 2.09 and 2.17 in the octahedral layer, the second (the Lieutenante smectites) with a sum close to 2.50, showing intermediate characteristics, at once di- and trioctahedral.

But in all cases, Mg is the preponderant cation of the octahedral layer, being higher than Al 4 times on 6. This magnesian character increases again in the phyllites derived from the trachydolerite.

This chemical composition is shown in the Al-Fe-Mg and Si-Mg diagrams (Fig. 2). The mineral issued from the trachydolerite (d1, d2, d3) occupy a different area from the others (a, b and c).

Otherwise, the 2/1 clay-minerals being identified as smectites, it is possible to calculate the *a* and *b* parameters from their classical analysis. The *b* parameters are included between 8.9 and 9.02 Å (Tab. 6). The ones, lower than 9 Å for the smectites derived from granite, diorite and basalt characterize montmorillo-

Table 4: Mineralogical composition of clay fraction of saprolites ($< 2\,\mu m$).

PARENT ROCK	GRANITE	DIORITE	BASALT	TRACHYDOLERITE	
	Moulin Blanc	Prignonet	Col du Mistral	La Lieutenante	
	a	b	c	d1	d2
% clay fraction	6.0	2.0	11.0	3.0	3.0
Apatite		0.30	0.40	1.00	1.00
Calcite		3.00			
Goethite	0.60	5.00	6.20	7.20	7.00
Illite	74.00	33.00	5.00	23.00	22.00
Kaolinite	-	15.00	20.00	0.50	6.00
Smectite	24.00	42.00	69.00	65.00	65.00
TOTAL	98.60	98.30	100.60	96.70	101.00

Table 5: Structural formulae of smectites - Bases: 11 oxygens.

SAMPLE	a		b		c		d1		d2		d3	
	Moulin Blanc		Prignonet		Col du Mistral		La Lieutenante					
	GRANITE		DIORITE		BASALTE		TRACHYDOLERITE					
Si'	4.03		4.02		4.00		3.75		3.71		3.40	
Al							0.25		0.29		0.60	
Al	1.034	2.17	0.700	2.16	0.765	2.09	0.688	2.488	0.875	2.472	0.630	2.552
Fe^{3+}	0.150		0.305		0.454		0.040		0.034		0.297	
Fe^{2+}	0.104		0.211		0.060		0.360		0.320		0.015	
Ti	0.010		0.060		0.080		0.070		0.063		0.015	
Mn'	0.080		-		0.100		-		-		0.028	
Mg	0.796		0.880		0.636		1.330		1.180		1.567	
Ca	0.100	0.206	0.190	0.253	0.200	0.280	0.057	0.317	0.051	0.273	0.204	0.334
Na	0.106		0.063		0.080		0.260		0.222		0.100	
K											0.030	
Charge												
Tetrahedral layer	+0.120		+0.080		-		−0.250		−0.290		−0.600	
Octahedral layer	−0.448		−0.563		−0.431		−0.456		+0.009		+0.061	
Interlayer	+0.306		+0.443		+0.480		+0.374		+0.324		+0.538	

Results extracted from the thesis of Chevalier (1984) - p.225

Remark: The d3 smectite of the Lieutenante with a strong substitution of Si by Al in the tetrahedral layer is close to the Beidellite of Velka Kopan - Carpathian Ukraine-Deer (1964).

Table 6: Analytical characteristics of smectites.

SAMPLE	a	b	c	d1	d2	d3
$Al_2O_3+Fe_2O_3/MgO$ ratio (1)	2.02	1.7	2.95	0.95	1.3	1.3
Parameters (Å) (2)						
a	8.977	8.975	8.979	9.009	9.008	9.023
b	5.183	5.181	5.184	5.200	5.200	5.209
Calculated surface (m^2/g)	706	720	719	720	727	727
Calculated CEC (me/100g) (3)	65	89	96	76	66	108

References: (1) Chanret et al. (1971), (2) Brown (1961), (3) Foster (1951).

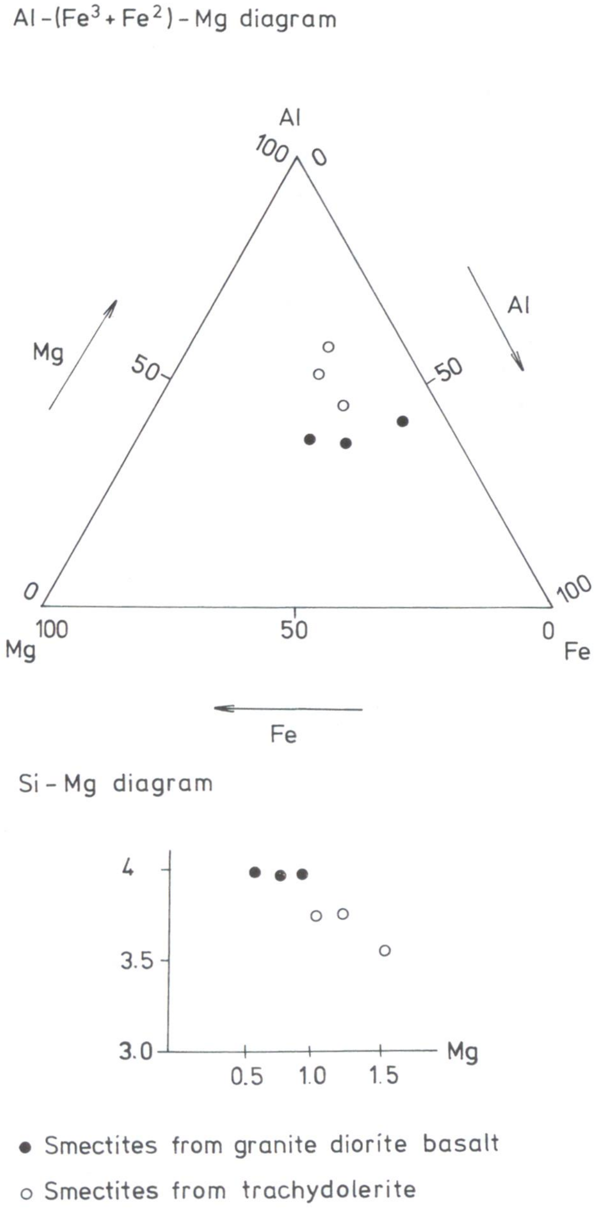

Fig. 2. Major elements of the octhohedral layer of smectites.

nites (Mac Ewan 1951). They are slightly higher than 9 Å for the smectites derived from the trachydolerite in which Si is partially substituted by Al in the tetrahedral layer, as in clay minerals close to the beidellite group (Nagelschmidt 1938).

The total surfaces vary only between narrow limits: 706 to 726 m^2/g.

The calculated CEC show a great variation, two among those being relatively weak (a and d2). But it is possible that an analytical investigation should give different results. On the contrary, the CEC of the c and d3 samples (96 and 108 me/g) are classical data for the smectites.

Discussion and conclusion

The structural formulae have shown the presence of two groups of smectites. The first one is formed by aluminous montmorillonites, rich in Mg, without Si-Al substitution in tetrahedral position and for which the Al_2O_3 + FeO / MgO ratio, very weak, relates those to Cheto montmorillonites (Chantret et al. 1971). The Li-test confirms this diagnosis. The second group, derived from the trachydolerite, has a different crystallographic structure, because there is a partial substitution of Si by Al in the tetrahedral sheet, while the sum of cations in the octahedral sheet varies from 2.47 to 2.55; it is also clearly higher than 2, but lower than 2.65, accepted limit for a trioctahedral layer (Foster 1960). Otherwise, the Al_2O_3 + FeO / MgO ratio varies from 0.95 to 1.3 characterizing magnesian smectites (Chantret et al. 1971), and this element is more abundant than in the first group and more important than Al in octahedral position. However, these smectites are not saponites, richer in Mg, but for which the number of cations R^{3+} in the octahedral position is always higher than 0.50 while the number of cations R^{2+} on the same position is lower than 85 % (Weaver & Pollard 1973).

Consequently, it seems that these clays are aluminous transition smectites, but especially magnesian, with a substitution of Si by Al like in beidellites and di-trioctahedral characteristics. This fact is corroborated by the behaviour of ATD curves. Their structural formulae classifies them close to magnesian type Md of Chantret et al. (1971), but similar to ferriferous beidellites, because the Si-Al substitution in the tetrahedral position and the Li-test. They are close to smectites derived from pillow-lavas of Cyprus for which the sum of cations in the octahedral site is 2.63 (Desprairies & Lapierre 1973). Such phyllites with an intermediate composition have been described by numerous authors (Colin et al. 1980; Curtin & Smillie 1981; Brigatti & Poppi 1982; Nahon et al. 1982; Macaire et al. 1988; Eggleton & Wang 1991).

However, relating to these transition-smectites, it is not possible to exclude the hypothesis of an intimate mixing of di- and trioctahedral smectites, like Graig & Loughman (1964) have demonstrated.

Relating to the difference of crystallographic structure of the two groups of the smectites, it is possible to call up the effect of leaching. The kaolinite fraction, weak among the alteration products of the trachydolerite, would speak in favour of a less active leaching than in the other sites of Provence with for consequence the genesis of an octahedral layer of the smectites better provided with movable cations: magnesian and iron.

Correlation between the chemical composition of parent rocks and the smectites derived from them

The genesis of neoformed smectites during the weathering of crystalline rocks of the Provence is joined, if all conditions are ever supposed, to the chemical and mineralogical compositions of the parent materials.

1 - The proportion of smectites in the < 2 μm fraction increases with the basicity of the parent rock, reaching more than 60 % in the saprolites derived from the basalt and the trachydolerite. This fact is in good agreement with the conclusions of Harder (1969), who shows that Mg, increasing regularly from the granitic range to the basic trachydoleritic one, facilitates the synthesis of smectites. This fundamental effect of Mg has been also shown by Güven & Pease (1975) and by Moinereau (1977).

2 - Concerning the chemical composition, Fig. 3 a, b show that the Provence smectites become more and more magnesian in succession with the increase of basicity of the parent rock. This element, leached from the primary ferro-magnesian networks: biotite, hornblende, olivine, is partially concentrated in the octahedral layers of smectites and also of illites, but especially in those of clayed 2/1 minerals formed in the basic rocks.

3 - The total iron (Fe_2O_3 + FeO) increases progressively from the granite (0.78 %) to the trachydolerite (7.85 %). It is the same in the smectites, however except for the trachydolerite. Otherwise, Fe enters in the composition of goethite which increases regularly in the fraction, as the basicity of the rock increases.

4 - In the end, the curves showing the evolution of the SiO_2 / bases ration in the rocks and in the 2/1 clays have similar trends.

Therefore, this population of smectites with different minerals, according to the environment in which they were formed, is the reflection of the genetic heritage of parent rocks.

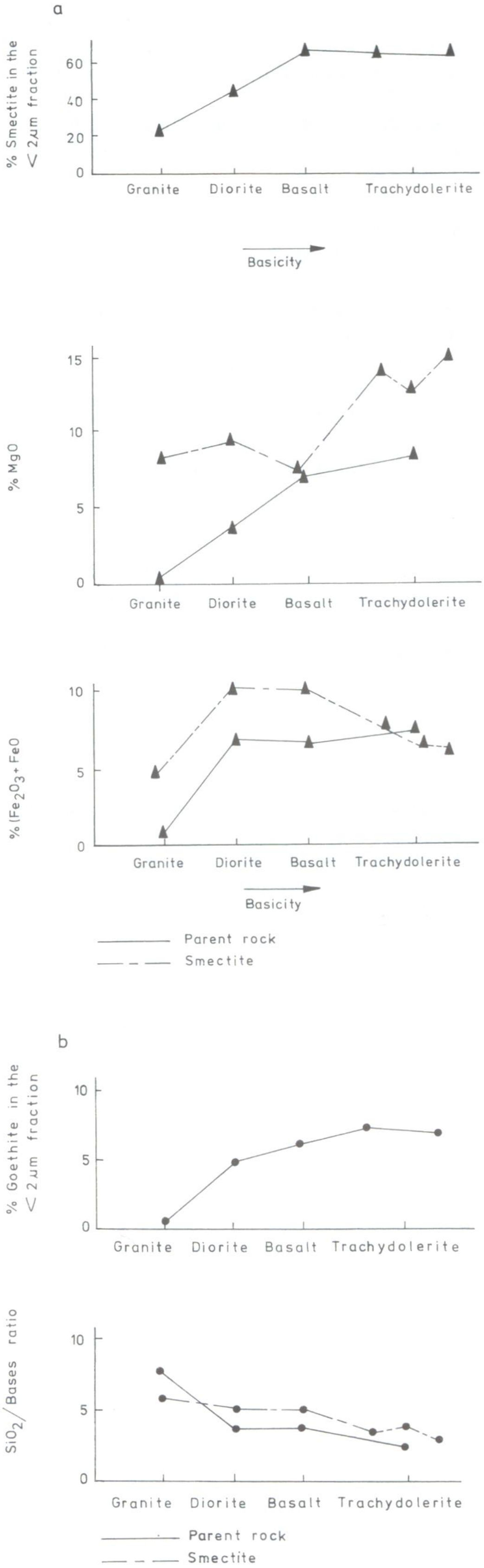

Fig. 3 a, b. Correlation between the chemical composition of the parent rocks and the derived smectites.

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