

CLAY MINERALOGY, DIAGENESIS, PALEOTHERMOMETRY AND HYDROCARBON POTENTIALS OF CRETACEOUS SHALES IN THE SOUTHERN BENUE TROUGH, NIGERIA

SAMUEL O. AKANDE¹ and ISTVÁN VICZIÁN²

¹Department of Geology and Mineral Sciences, University of Ilorin, P. M. B. 1515, Ilorin, Kwara State, Nigeria

²Hungarian Geological Survey, Stefánia út 14, Budapest H-1143, Hungary

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Abstract: In the axial zone of the Abakaliki Basin (southern Benue Trough, Nigeria), Middle Cretaceous shales have undergone normal regional burial which is superposed by Santonian tectonism and local heating effects around hydrothermal centres (Ishiagu, Enyigba, Ameri). Illite crystallinity, IC ranges from 0.4 to 0.9 °2 Θ , smectite proportion in mixed-layer illite/smectites, S = 0 % and reflectance of vitrinite, Rm ranges over broad limits of 2.46 to 4.30 %, all indicating strong diagenetic to anchimetamorphic transformations. The Upper Cretaceous shales outside the axial zone and in the Anambra Basin have only been exposed to normal burial diagenesis in areas around Ezillo, Ihe and Leru-Okigwe. In these locations, IC = 1.4 to 1.6 °2 Θ , S varies from 20–30 % and Rm between 0.7 and 0.9 %. Paleotemperature estimates are up to 230 °C in the axial zone and about 100 to 140 °C in the marginal areas. The fairly mature Late Campanian Enugu/Nkporo Shales are possible source beds for liquid petroleum whereas the Turonian Eze-Aku Shales are more gas prone. The Albian Asu River Shales are overmature and contain gas prone organic constituents.

Key words: Cretaceous, Nigeria, Benue Trough, clay minerals, diagenesis, hydrothermal alteration, petroleum source rocks.

Introduction

Stratigraphic relations and thermal effects around the mineralized zones in the axial region of the Benue Trough were reported in Akande et al. (1992). The paper contains vitrinite reflectance, organic petrology and fluid inclusion data which constrained the grade of mineral transformation, temperature of hydrothermal vein formation and igneous intrusive effects in the Benue Trough. Few illite crystallinity data in the neighbourhood of igneous intrusions were included in that paper. The objectives of the present study is to add to the geothermometric methods used so far by investigating the degree of illite crystallinity and smectite contents of mixed-layer illite/smectites in the shale sections of the axial region and the marginal areas of the southern Benue Trough (Fig. 1) and to interpret these in the light of some previous data on organic petrology, burial diagenesis and petroleum source rocks.

Regional stratigraphic setting

The study area forms a part of the southern Benue Trough comprising of the Abakaliki and the Anambra Basins. It is underlain by a north-east trending belt of Cretaceous sediments the evolution of which resulted from block faulting, basement fragmentation and rifting during the Early Cretaceous opening of the South Atlantic ocean. Sedimentary infill of the southern Benue Trough consists of three major unconformity bounded depositional cycles: the Albian-Cenomanian, Turonian-Coniacian and the Campanian-Maastrichtian cycles (Petters

1977). In the Abakaliki Basin, these sequences consist of the Aptian-Albian Asu River Group, Eze-Aku Formation (Cenomanian-Turonian), Awgu Formation (Coniacian) and in the Anambra Basin, post Santonian sediments include the Enugu/Nkporo Formation (Late Campanian) and the Mamu Formation (Maastrichtian) (Fig. 1). The Cretaceous sediments reached a thickness of more than 5000 m of which over 2000 m were deposited in the Anambra Basin (Petters 1977; Agagu & Adighije 1983) in the Campanian to Maastrichtian times. Sediments of the Asu River Group, the Eze Aku and the Awgu Formations were exposed in the core of the Abakaliki anticline and in the areas marginal to it while the overlying Nkporo and Mamu Formations occur within the adjacent Anambra Basin (Fig. 1). A detailed stratigraphic description of the Benue Trough is presented elsewhere and will not be discussed further here.

Materials for study

Sample materials for this study were selected over a wide range of stratigraphic intervals (Fig. 1, Tab. 1). In the axial part of the Lower Benue Trough (i.e. the Abakaliki anticline), Albian shales of the first depositional cycle were collected at Ishiagu, Enyigba and Ameri where open cast mining activities have exposed the successions of the Aptian-Albian Asu River Shales. Surface samples of shales of the Eze-Aku Formation; Turonian (second depositional cycle sediments) were collected at Ezillo and those of the Late Campanian Enugu/Nkporo Shales (third depositional cycle) were col-

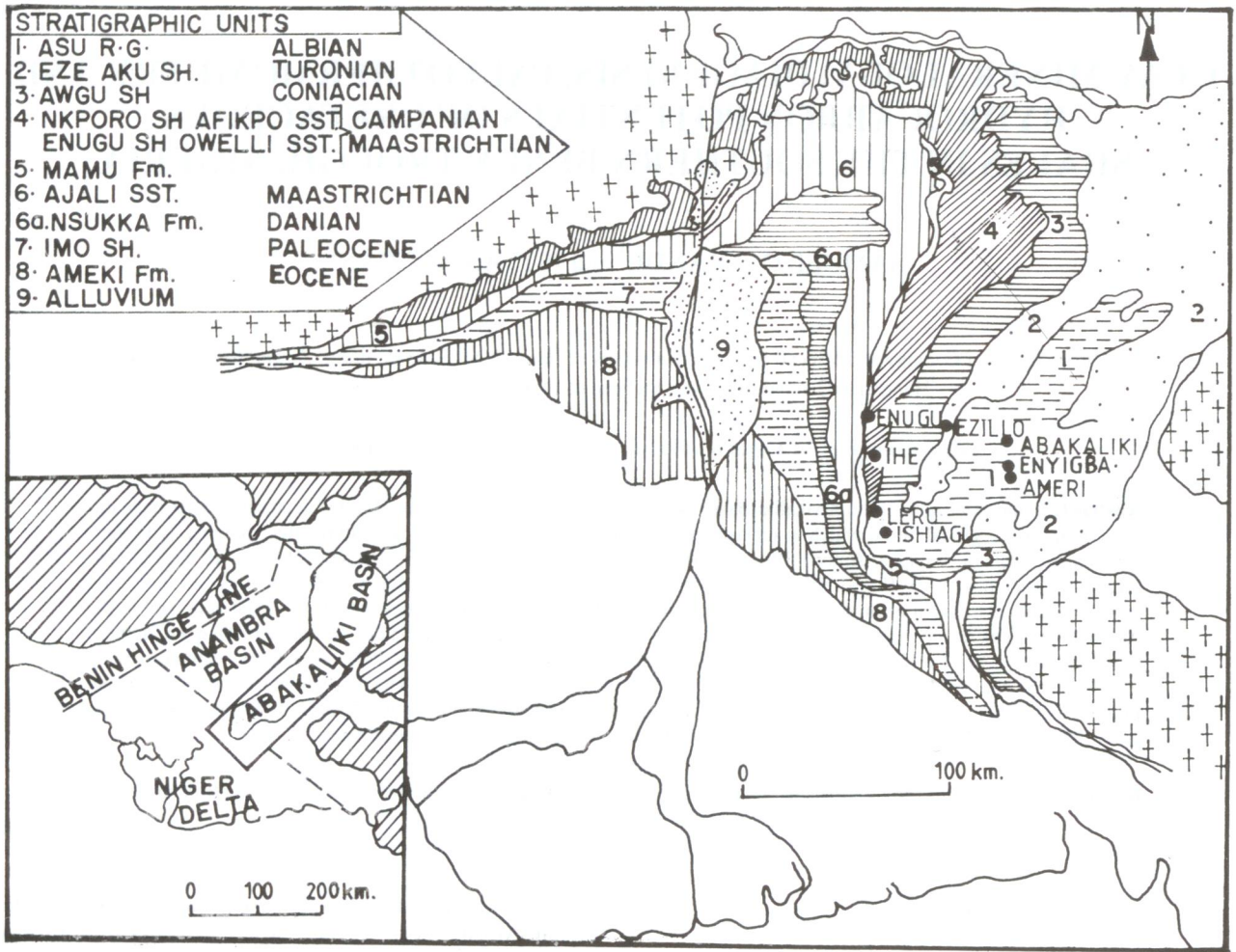


Fig. 1. Geological map and megatectonic framework of southern Nigeria (inset map). Sample locations are shown on the geological map.

lected at Ihe and Leru-Okigwe in the marginal areas of the Anambra Basin. A total of thirty two samples were collected at the sampling locations for both organic petrology and mineralogical analysis. Twelve of these samples representing a wide range of vitrinite reflectance R_m values were selected for detailed clay mineral analysis.

Macroscopic features of the samples selected for clay analysis (e.g. colour, petrography, textures and structures) are described briefly in Tab. 1. The rocks are generally massive or laminated shales with variable carbonate contents.

Methods

Sample preparation, X-ray methods

The samples were crushed to few millimeter size. Part of the crushed material was powdered and used for the X-ray analysis of the bulk samples. The other part was treated with 3% HCl in order to remove the carbonates. From the carbonate free materials, the fraction $<2 \mu\text{m}$ was separated by settling in Atterberg cylinder. X-ray analysis of the bulk samples was carried out on non oriented, powder specimens. From the $<2 \mu\text{m}$ fractions, oriented samples were made by the smear on

glass method. X-ray patterns were taken of the untreated and of the ethylene glycol treated material. X-ray patterns were made using $\text{CuK}\alpha$ radiation and graphite monochromator. Smectite proportions (S) and degree of ordering of the component layers in mixed-layer illite/smectites of the $<2 \mu\text{m}$ fraction were determined by the method of Šrodoň (1980). The position of the basal reflections in the $15\text{--}18^\circ 2\theta$ and in the $44\text{--}45^\circ 2\theta$ ranges and the angular distances defined by Šrodoň as Δd_1 and Δd_2 were measured on the ethylene glycol treated pattern (Fig. 2). Only 4 samples contained smectite proportions in the mixed-layer structure (Tab. 2).

Illite crystallinity is defined as the width of the 10 \AA illite reflection at half height (IC, Kübler 1964, 1990). IC values were measured from air-dry specimens, both of the bulk samples and of the $<2 \mu\text{m}$ fractions. From the $<2 \mu\text{m}$ fraction, two oriented specimens were measured for each sample. The angular range of $4\text{--}10^\circ 2\theta$ was recorded with $1/2^\circ 2\theta/\text{min}$ goniometer speed using step scanning with 0.01 step size, 1 s counting interval, 11 point smoothing and background correction. Background correction is a crucial point because the reflections with the d values around 10 \AA may be influenced by the steep background due to the rise of the Lorentz polarisation factor towards low θ angles. Typical line profiles of the 10 \AA illite reflection are shown in Fig. 3. The average difference between

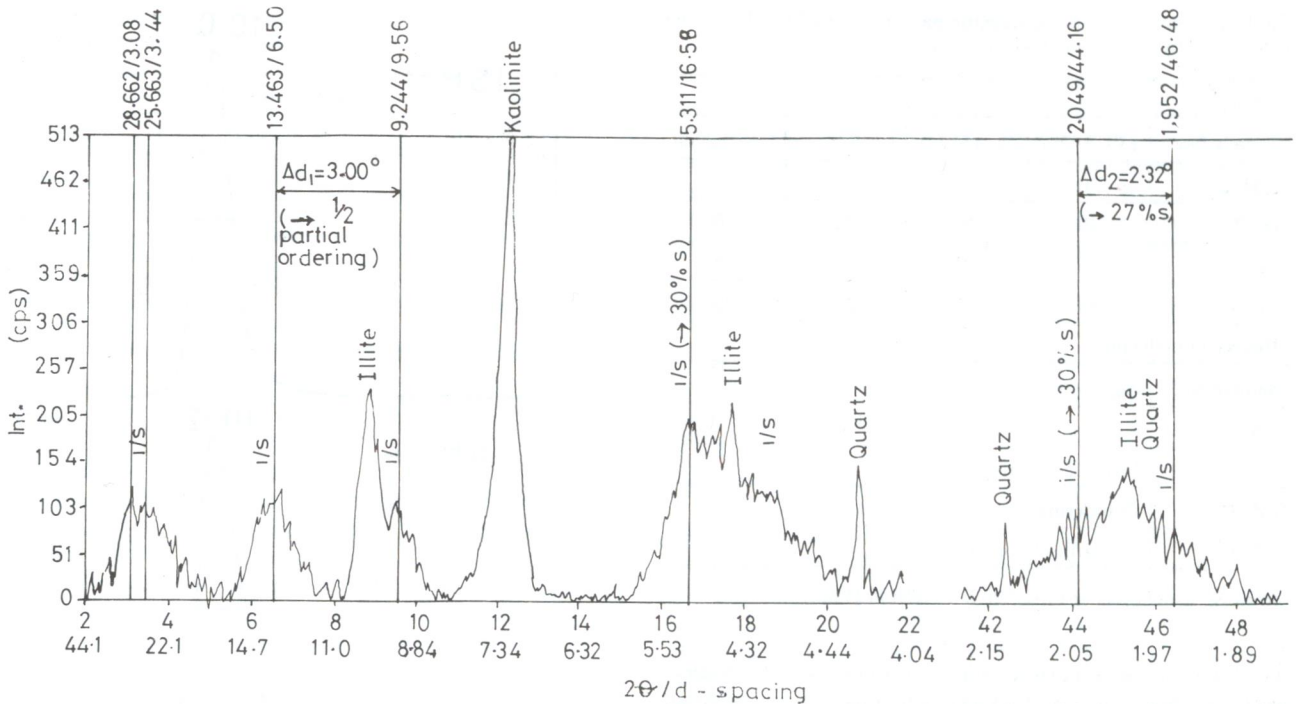


Fig. 2. Illustration of the application of the Šrodoň (1980) method for the determination of the smectite proportion (S) and degree of ordering in mixed-layer illite/smectites. X-ray diffraction pattern of sample EZ-4, <2 μm fraction, treated with ethylene glycol. Instrumental setting is described in the text. The method of measuring the angular distances Δd_1 and Δd_2 is shown. I/S: basal reflections of illite/smectite.

the measurements on two oriented specimens prepared from the same sample is $0.04^\circ 2\theta$.

The position of maximum of the first order basal reflection, $d(001/001)$, is another parameter depending on the ratio of discrete illite to mixed-layer illite/smectite. In the case of the present study it varies from 10 \AA (dominantly illite) to 11.5 \AA (illite/smectite). Examples are shown in Fig. 3. The average difference between two parallel measurements is 0.03 \AA for il-

litic material and 0.15 \AA for samples containing mixed-layers. Because of uncertainties arising from the orientation effect, no quantitative determination of all the mineral phases present in the <2 μm fraction was attempted. Only clay minerals were compared with each other. Their proportion was estimated by multiplying the integrated intensities of the basal reflections by suitable factors. Numerical values of these factors were discussed by Rischák & Viczián (1974). The presence of other phases in the <2 μm fraction is only indicated. For the bulk samples investigated in randomly oriented powder specimens, quantitative estimation of all phases was made. The quantitative estimation is based on the direct comparison of reflection intensities of the phases present, a modified version of the method described by Viczián (1967), normally used in the X-ray laboratory of the Hungarian Geological Survey, Budapest.

Calibration of the measured IC values with the international scale

The data of IC obtained by individual laboratories cannot be directly related to diagenesis and anchimetamorphism because of the differences in sample preparation, instrumental setting etc. among laboratories. In order to calibrate the data set with the internationally accepted standard scale, three samples were kindly measured by Dr. P. Árkai in the Geochemical Research Laboratory of Hungarian Academy of Sciences, Budapest. This enabled us a direct comparison with the original standards of Kübler. The results are shown in Tab. 3. IC values measured by us are slightly higher than those of Árkai but there is a relatively good linear relationship. By means of the calibration, measured IC values were converted to the International scale (by multiplying with 0.88).

One other problem is the definition of the boundaries of the anchizone in terms of IC values. According to Kübler (1990),

Table 1: Description of samples used for clay mineral analysis.

Sample No.	Location	Description	Age
Abakaliki anticline (axial zone)			
ISH-1	Ishiagu	Grey laminated shale, sideritic	Albian
ISH-2	Ishiagu	Grey laminated shale, sideritic	Albian
ISH-3	Ishiagu	Grey laminated shale, sideritic	Albian
ISH-4	Ishiagu	Dark grey shale, calcareous	Albian
ISH-5	Ishiagu	Grey laminated shale, sideritic	Albian
ENY-8	Enyigba	Grey laminated shale, sideritic	Albian
ENY-9	Enyigba	Grey shale, sideritic	Albian
ENY-10	Enyigba	Grey shale, calcareous	Albian
AM-2	Ameri	Shaly limestone	Albian
Anambra Basin (marginal areas)			
EZ-3	Ezillo	Silty shale, finely laminated	Turonian
EZ-4	Ezillo	Silty shale, finely laminated	Turonian
IH-2	Ihe	Silty shale, finely laminated	Late Campanian

Table 2: Comparison of the smectite proportions (S%) and ordering determined by various methods of Środoń (1980).

S%				
Sample No.	15-18 °2θ	44-45 °2θ	Δd ₂	Conclusion
AM-2	20	-	-	20
IH-2	20	20	20	20
EZ-3	26	30	20	25
EZ-4	30	30	27	30
Degree of ordering				
Sample No.	Δd ₁			
AM-2	-			
IH-2	ISII - IS ordering			
EZ-3	IS ordering			
EZ-4	1/2 partial IS ordering			

Remark: using plots of Figs. 5 to 8 in Środoń (1980)

the limits of the anchizone are 0.25 and 0.42 °2θ, respectively. There are, however, other definitions, e.g. that of Árkai (1987), who defines the anchizone between the limits 0.20 and 0.30 °2θ in the same scale. The difference in the definition is explained by Árkai as caused by the differences in the lithology (especially in the contents of detrital white micas) of the representative areas and by the necessity to apply a complex set of parameters instead of the single parameter IC.

In the cases studied by Árkai (1987) the lower IC values for the limits of the anchizone are consequences of the detrital micas even in the <2 μm fraction. This is, however, not the case in the Nigerian samples (see Tab. 4). Therefore, in the present study the limits defined by Kübler were accepted.

Results

The mineralogical composition and parameters characterizing the clay minerals are given in Tab. 4 for bulk mineral samples and in Tab. 5 for the clay fractions. Sample locations can be grouped into two regional and structural units i.e. those from the axial zone of the Abakaliki anticline and those from the marginal areas.

The axial zone

The set of samples from the axial zone of the Abakaliki anticline (Ishiagu, Enyigba and Ameri) are from the Albian Asu River Formation:

In the samples from Ishiagu (samples ISH-1, -2, -3 and -5) the only clay mineral is illite. Carbonate contents (predominantly siderite) in these samples vary from 16 to 25 %. Siderite seems to be well ordered having nearly ideal composition with no apparent isomorphous substitution. Illite is predominantly 1Md which is the typical polytypic modification of neoformed illites in the diagenetic zone. The modification 2M is much less abundant. 2M represents inherited detrital micas. Also the <2 μm fraction contains the single clay mineral illite. The sample ISH-4 has somewhat different composition having high calcite and low siderite contents. IC in this sample is somewhat higher than elsewhere in Ishiagu (0.65 °2θ). All the samples contain pyrite which results in the grey

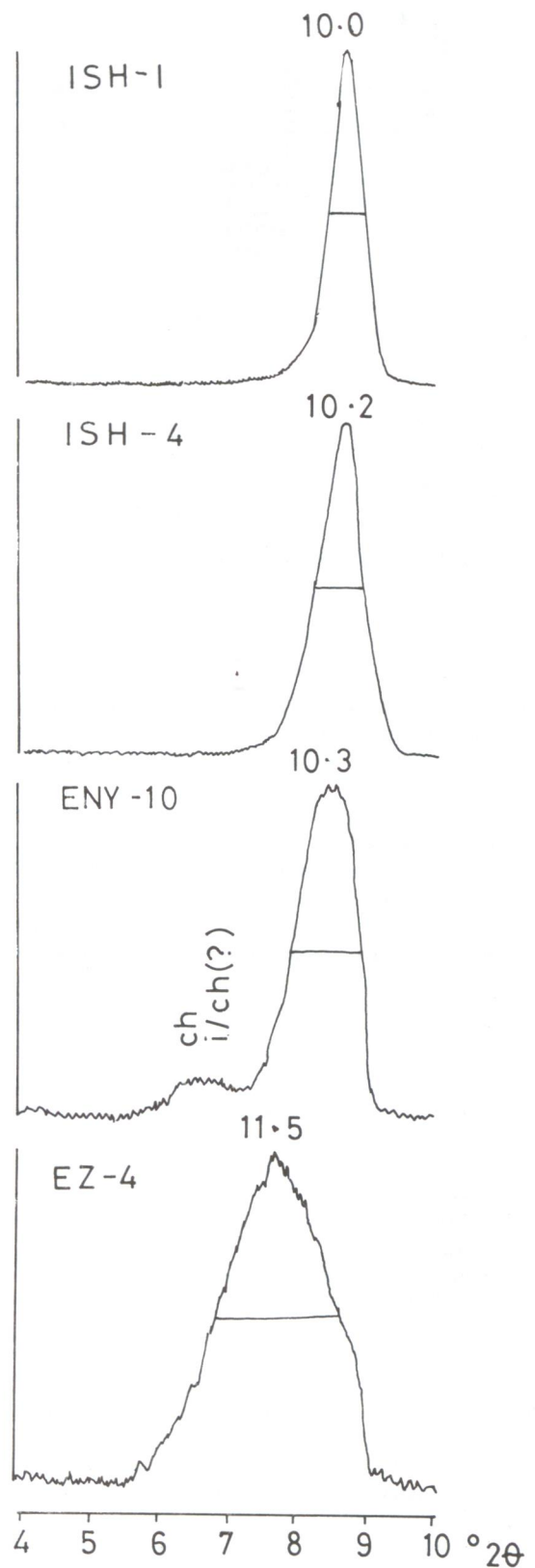


Fig. 3. Typical line profiles of the first order illite (or illite/smectite) basal reflection showing variation of the width at half height (illite crystallinity, IC) and the d value of the peak maximum (Å), <2 μm fraction, untreated specimens. Instrumental setting is described in the text, ch: chlorite, i/ch(?): illite/chlorite(?).

Table 3: Calibration of measured illite crystallinity (IC, °2 θ) values with the internationally accepted scale.

Sample No.	Viczián, HGS	Árkai, GRL
ISH-1	0.49 ± 0.00	0.42 ± 0.004
ISH-4	0.74 ± 0.01	0.62 ± 0.015
ENY-10	0.98 ± 0.02	0.88 ± 0.013

HGS: Hungarian Geological Survey, Budapest

GRL: Geochemical Research Laboratory of Hungarian Academy of Sciences, Budapest

All measurements were made with 1/2 °/min goniometer speed.

colour of shales. Anatase was identified in the diffractograms by the reflection at $d = 3.52 \text{ \AA}$.

At Enyigba, the samples ENY-8 and -9 are similar to the illitic and sideritic shales of Ishiagu, however, IC values (0.62 and 0.65 °2 θ) are somewhat higher than those at Ishiagu. Sample ENY-10 is similar to sample ISH-4 having high calcite and low siderite contents. Its special oxidised character is shown by a high goethite content which is obviously concentrated in the observed reddish brown concretions with yellow rim. Another special feature of this sample is the chlorite content and the presence of a mixed-layer 10/14 Å clay mineral. This is probably illite/chlorite because no shift of the position of the basal reflection was observed upon glycolation (see Fig. 3).

At Ameri, the third locality in the axial zone, the sample AM-2 is a high calcite-bearing shaly limestone. Siderite is absent but the sample contains an iron-rich chlorite which is the dominant mineral in the <2 μm fraction. High iron content in the chlorite is shown by the high 002 and 004 and the weak 001 and 003 basal reflections (see Fig. 4). The crystallinity of illite (IC) in this sample is 0.38 °2 θ (the lowest crystallinity recorded in the group). It is noted, however, that this sample also contains some mixed-layer illite/smectite which is not sufficient enough to influence the basal reflection of illite at half peak height. At slightly higher contents of illite/smectite the IC would increase to 0.6 or 0.7 °2 θ .

The marginal areas

The second set of samples from Ezillo and Ihe are located west of the Abakaliki anticline. At Ezillo the samples EZ-3 and EZ-4 were collected from the Turonian Eze-Aku Shales. Sample from Ihe (IH-2) is from the Late Campanian Enugu Shales. Shales in the two localities are dark grey, laminated, silty and carbonate free. Both the bulk rock samples and the <2 μm fraction contain the same clay minerals: kaolinite, illite and mixed-layer illite/smectite. Kaolinite is medium well ordered. Width of 001 reflection at half height ranges from 0.40 to 0.53 °2 θ (<2 μm fraction, for comparison see e.g. data collected by Rischák & Viczián 1974, see Tab. 5). The low-angle reflection of overlapping illite and illite/smectite is very broad: IC values are between 1.36–1.57 °2 θ . Even the maximum of the peak moves from exactly 10 Å towards higher d values (up to 11.5 Å) showing the predominance of mixed-layer illite/smectite over discrete illite (see Fig. 3, sample EZ-4). Smectite proportion in mixed-layer illite/smectite (S) varies between 20 and 30 % (see Fig. 2).

Discussion

The IC values and smectite proportions in mixed-layer illite/smectite (S) in the <2 μm fraction are plotted as a function of Rm in Fig. 5. Both IC and S vary with Rm and increasing diagenetic grade in a similar manner, they decrease with increasing Rm. S reaches its minimum (S = 0 %) at about Rm = 2.5 % and its value remains 0 % with further increase of Rm. Also IC decreases less rapidly at higher values of Rm.

Variation of IC, S and Rm with the bulk mineralogy

Three compositional groups can be distinguished on the basis of the bulk mineralogical assemblages: (1) silty shales, (2) calcareous shales and (3) sideritic shales.

1 — Silty shales:

The less transformed silty shales (samples EZ-3, -4 and IH-2) occur west of the Abakaliki anticline. The samples contain normal sedimentary polymineralic clay mineral association, S = 20–30 % smectite proportion in illite/smectites, high IC values of about 1.5 °2 θ and low Rm values in the range 0.7–0.9 %. These shales were affected by a normal burial diagenesis and are undeformed. They clearly belong to the zone of diagenesis on the basis of the classification of Kübler. Eberl (1993) pointed out the possibility of the subdivision of the diagenetic illitization processes into 3 zones. From Eberl's (1993) classification, these rocks are presently in the lower part of Zone 2 of illite formation where illites are thought to precipitate at the expense of the dissolution of K-feldspar and smectite. The Rm values of these shales suggest that they are in the oil generation zone (Tissot & Welte 1978).

2 — Calcareous shales:

These samples (ISH-4 and ENY-10) occur closer (about 2 km) to known intrusive body or hydrothermal veins of the Ishiagu and Abakaliki districts in the axial part of the Abakaliki anticline. The two samples have no observable smectitic mixed-layering in the illites, and IC values are slightly higher than those of other samples from the same locality (0.65 and 0.86 °2 θ , respectively). These may be due to weaker heat effect and also to the protecting effect of the calcitic matrix around the illitic particles that prevents the flow of solutions. They can be classified in the Kübler scheme as diagenetic. The IC data and Rm values (2.46 and 2.66 %) point to a diagenetic transformation grade near to the boundary of anchimeta-morphism. These two samples have the lowest Rm values in the two districts because of their relatively distant position from heat sources.

A shaly limestone sample (AM-2) from Ameri in the Abakaliki district consists of an iron-rich chlorite. Although the sample contains smectite in the mixed-layer phase, the IC value of 0.38 °2 θ from this sample is the lowest in the study areas. Similarly, the Rm value of the sample (3.52 %) suggests anchimeta-morphic conditions (Kisch 1987) beyond the zone of dry gas generation (Tissot & Welte 1978).

3 — Sideritic shales:

These samples (ISH-1, -3, -5, ENY-8, -9) are found at hydrothermal centres where vein type mineralizations or igneous intrusions occur in the Ishiagu and Enyigba districts. The

Table 4: Semi-quantitative mineralogical composition (%) of bulk rock samples.

Sample No.	r.?	i/s	i	k	ch	q	an	kfp	pl	c	d	si	py	g	am	illite polytypism
ISH-1			37			43	2					17	1			1Md>2M
ISH-2			34			39	1					25	1			1Md>2M
ISH-3			32			48	2					17	1			1Md>>2M, 1M(?)
ISH-4			9			20	1			62	4	3	1			1Md
ISH-5			33			48	2					16	1			1Md>>2M
ENY-8			37			52	2	tr.				7	2		tr.	1Md>2M>1M(?)
ENY-9			29			52	1					18				1Md>2M>1M
ENY-10	tr.	4	8		12	29				34		3		10		1Md>2M(?)
AM-2		2	5		16*	20				62			1	tr.		
IH-2		11	13	41		30	2	3								
EZ-3		32	17	17		22	3	2	7							
EZ-4		23	14	17		31	4	4	5	1			1			

* Fe-chlorite

r.: rectorite, i/s.: illite/smectite, i: illite, k: kaolinite, ch: chlorite, q: quartz, an: anatase, kfp: K-feldspar, pl: plagioclase, c: calcite, d: dolomite, si: siderite, py: pyrite, g: goethite, am: X-ray amorphous material.

Table 5: Semi-quantitative mineralogical composition (%) and characterization of clay minerals of the <2 µm fraction.

Sample No.	Rm%	i/s+i%	i%	k%	ch%	q	an	g	d(001/001), Å	IC, °2θ	kaolinite B(001), °2θ
ISH-1	4.27		100			+			10.0	.43	
ISH-2	4.25		100	tr.		+			10.1	.46	
ISH-3	3.22		100			+	+		10.1	.49	
ISH-4	2.46		100			+	+		10.2	.65	
ISH-5	2.73		100			+	+		10.1	.58	
ENY-8	4.03		100			+	+		10.2	.62	
ENY-9	4.30		100	tr.		+	+		10.1	.65	
ENY-10	2.66		93		7**	+		+	10.3	.86	
AM-2	3.52	36			64*	+			10.0 10.6	.38	
IH-2	0.67	55		45		+	+		10.5	1.36	.40
EZ-3	0.90	91		9		+	+		11.1	1.43	.53
EZ-4	0.90	88		12		+	+		11.5	1.57	.42

*Fe-chlorite, **discrete chlorite + mixed-layer illite/chlorite(?)

Rm values are taken from Akande et al. (1992).

Abbreviations of minerals see: Table 4.

Rm: vitrinite reflectance, d(001/001): position of the first basal reflection of mixed-layer illite/smectites, kaolinite B(001): width of the 001 reflection at half height.

sideritic shales occur proximal to hydrothermal veins with siderite, quartz, calcite or sulphide infillings. In this group of samples, the only clay mineral is illite. No smectite interlayering can be observed in illite (S = 0 %). Rm in the study areas vary within broad limits (2.73 to 4.30 %), depending on the intensity of the thermal upsurges which accompanied the hydrothermal mineralization. IC varies inversely with Rm within narrow limits (0.43 to 0.65 °2θ). All Rm values are high and indicate the anchizone according to Kübler's (1990) defini-

tion. The terms anchimetamorphism or anchizone are used here to express the degree of the organic maturation, however, the nature of the transformation differs from that observed in regional metamorphic terranes. The IC values remain in the zone of diagenesis (with values higher than 0.42 °2θ). The samples fall within the approximate boundary limit of the anchizone. This is probably due to the relatively short duration of the hydrothermal effects as compared with the time of heating during the regional burial metamorphism. Illite seems to

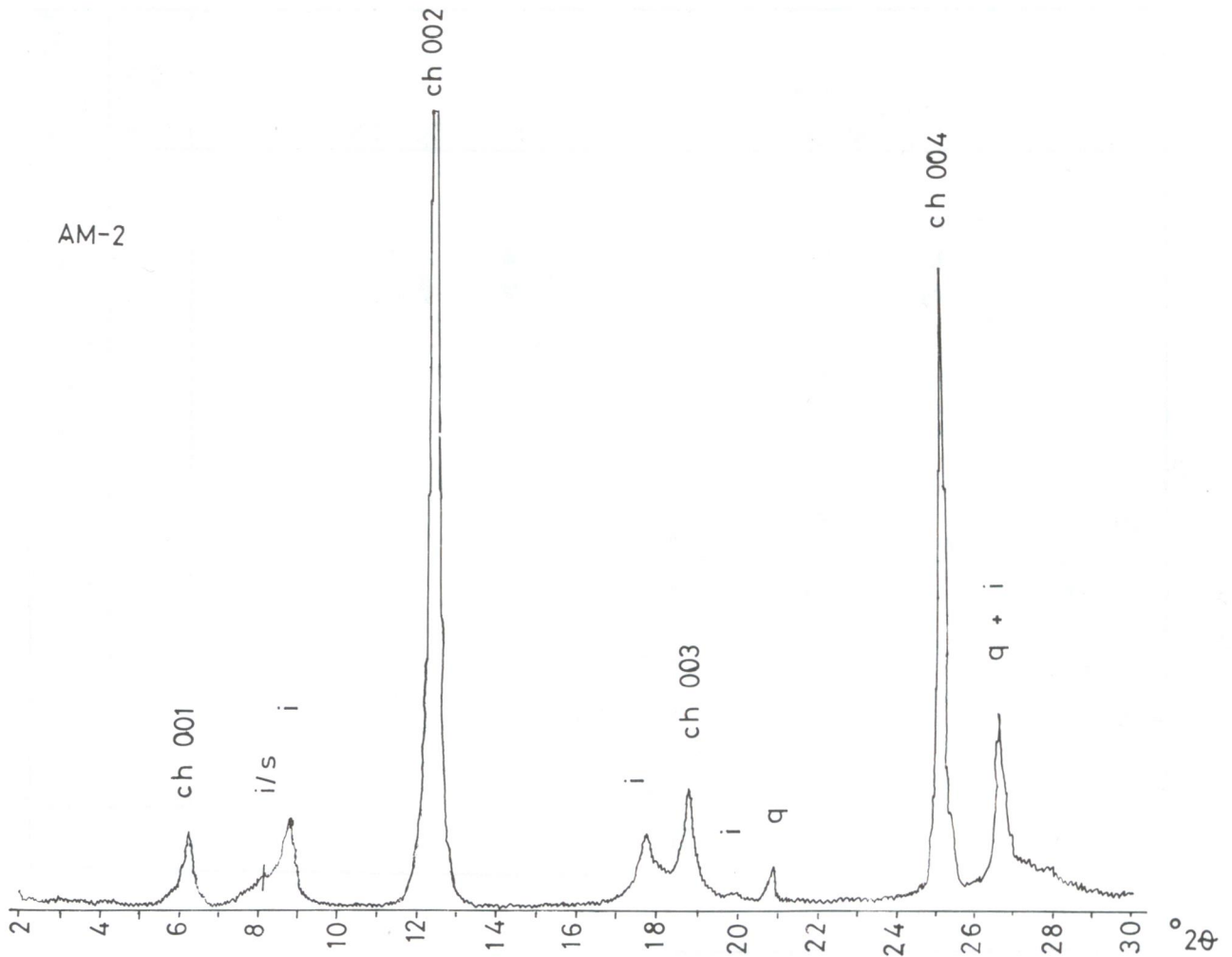


Fig. 4. X-ray diffraction pattern of sample AM-2, <2 μm fraction, untreated specimen. The relatively strong even order and weak odd order basal reflections of chlorite indicate Fe-rich chlorite (Petruk 1964). Instrumental setting is described in the text, ch: chlorite, i: illite, i/s: illite/smectite, q: quartz.

react to the heat effects at a slower rate than organic matter. All the Albian sediments (compositional groups 2 and 3) belong to the Zone 3 of diagenesis according to the zonation of Eberl (1993). The mechanism of illite transformation in this zone is attributed to coarsening of particles and perfection of crystal structure. This slow process is generally delayed compared to the transformation paths of organic matter (Kisch 1987, p. 246).

Another difference between the hydrothermal effect and regional metamorphism is shown in the polytypism of micas. Anchimetamorphic rocks usually contain the modification 2M (Maxwell & Hower 1967) while in the study area the dominant modification is 1Md (Tab. 4).

The non-equilibrium character of the illite transformation is shown also in the consistent difference between the IC values found at Ishiagu and Enyigba. In the published data of Akande et al. (1992), maximum temperatures, size of the thermal anomaly, lithology and the types of hydrothermal vein mineralization are similar in the two localities. Difference in the intensity of heating may be deduced from sphalerite homogenization temperatures which are about 15 $^{\circ}\text{C}$ higher at Ishiagu than at Enyigba (Akande et al. 1992). Another reason

for the somewhat higher grade of metamorphism at Ishiagu may be the additional heat effect of the dolerite dykes, sills and plugs which invaded the sideritic shales after the hydrothermal vein mineralization.

Estimation of paleotemperatures

Although the thermodynamic status of clay minerals is still a disputed subject, Lippmann (1982) seems to have convincingly proven that mixed-layer illite/smectites are metastable phases in a thermodynamic sense. As a consequence, there is no absolute relationship between smectite proportions (S) and temperature of formation of these minerals. There are, however, empirical observations indicating that for a given set of circumstances such a relationship can be established (Viczián 1985). Some examples are shown in Fig. 6 indicating a calibration of S with respect to temperature. Relatively low temperature data are given for the normal burial of older sediments. In rapidly subsiding younger basins temperatures are generally higher. This can be observed in the examples of the Plio-Pleistocene of the Colorado River delta (Jennings & Thompson 1986), the Neogene of the Vienna and Pannonian

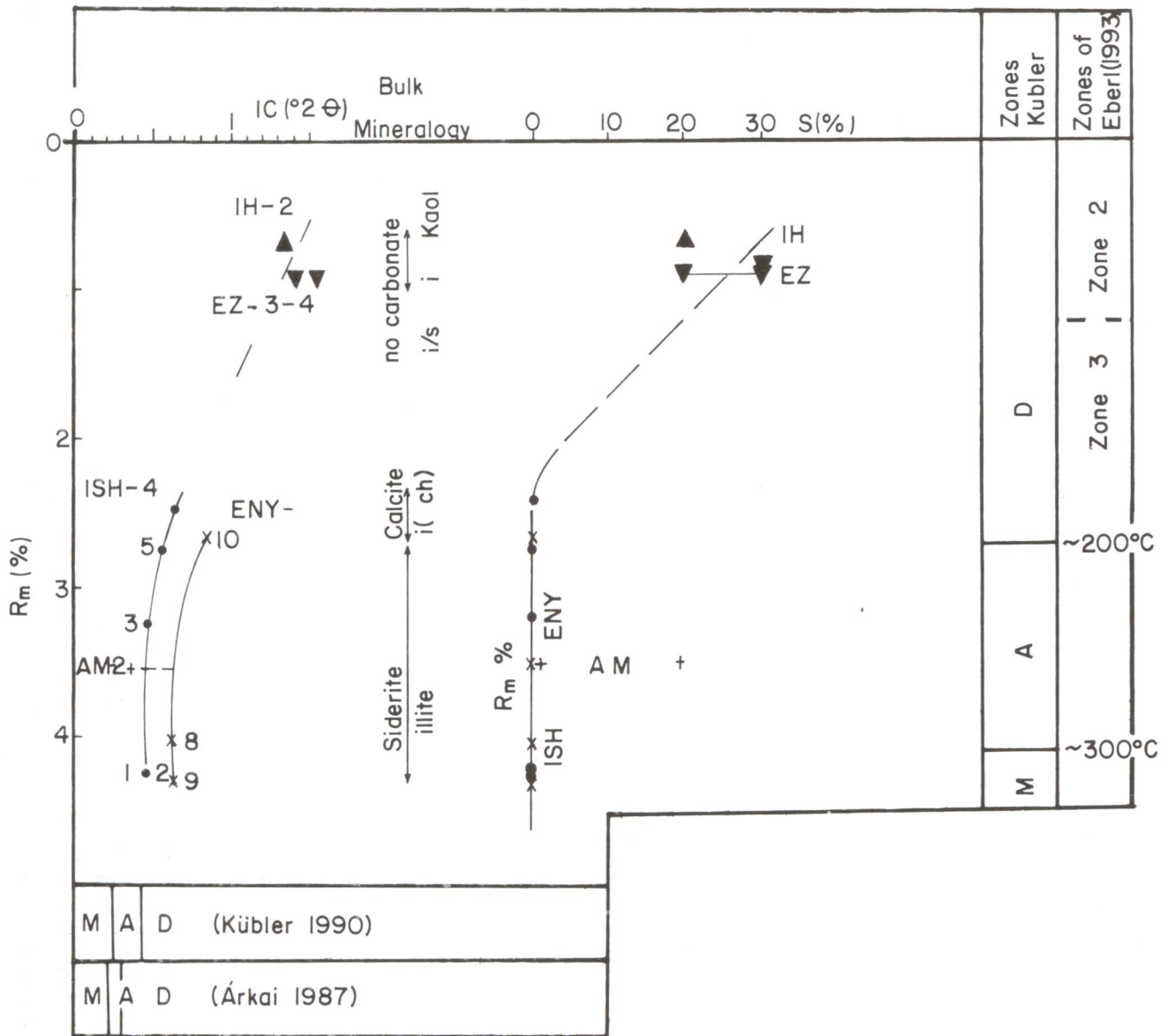


Fig. 5. Plots of illite crystallinity (IC) and smectite proportion in illite/smectites (S) of the <math><2\ \mu\text{m}</math> fraction versus vitrinite reflectance (R_m) in samples from the Benue Trough. The bulk composition of the samples is indicated. Abbreviations of the minerals are the same as in Fig. 4. Metamorphic zonation according to Kübler (1990) and Árkai (1987) is shown parallel with the IC axis, D: diagenesis, A: anchimetamorphism, M: metamorphism. Kübler's zonation based on R_m values as well as Zones 2 and 3 of illite formation according to Eberl (1993) are shown. The approximate limit between Zone 2 and 3 is drawn at $S = 20\%$. Notations of the boreholes (see also Tab. 1): \bullet ISH, \times ENY, $+$ AM, ∇ EZ, Δ IH.

Basins (Slovakia, Hungary, Francù et al. 1990; Šucha et al. 1993; Viczián 1994) and the Paleogene-Lower Miocene of the Gulf Coast (Hower et al. 1976) where temperatures may vary from 80 to 220 °C with corresponding decrease in the proportions of smectite from 40 to 10 %. Temperatures from 100 to 110 °C are deduced for the onset of the IS-type mixed-layering at about $S = 40\%$ in a review by Pollastro (1990). Based on these correlations, a temperature range of 100 to 140 °C was probably attained by the Turonian to Maastrichtian sediments in the western margin of the Abakaliki anticline (Ezillo and Ihe locations) and the Anambra Basin.

The groups of samples from the axial zone of the lower Benue Trough (Ishiagu, Enyigba and Ameri) were exposed to

a more complex thermal history than the marginal areas. Apart from the normal burial diagenesis, these samples have been affected by both thermal and tectonic events. A comparison of normally buried sedimentary sequences and short lived geothermal systems shows that in the latter case higher temperatures are needed to reach the same degree of illite/smectite transformation (Pollastro 1990, see Fig. 6).

The temperature range of the anchizone, estimated from R_m values falls between 200 and 300 °C (Árkai 1987). Kisch (1987) suggests 200–250 °C for the onset of anchizone. Akande et al. (1992) obtained 240 °C from fluid inclusion studies in the study areas and noted that higher temperatures and anchimetamorphic conditions prevailed at the centres of

S(%)	burial, PG-M Gulf Coast	burial, NG Vienna and Pannonian B.	burial, Pli-Q Colorado River delta	Pollastro (1990)	
				burial, (Δ 2 Ma)	Short-lived Geothermal Systems
			°C		
40	80		130	100- 110	130- 140
30	90	105	140		
20	110	140	155	175- 180	
10		180 220	170 190		
0			≥ 230		

Fig. 6. Dependence of smectite proportion in illite/smectites (S) in various sedimentary basins on temperature and heating time. Source of data: Gulf Coast: Hower et al. (1976), Vienna and Pannonian Basins (Slovakia, Hungary): Franců et al. (1990), Šucha et al. (1993), Viczián (1994), Colorado River delta: Jennings & Thompson (1986). PG: Paleogene, M: Lower Miocene, NG: Neogene, Pli: Pliocene, Q: Pleistocene.

hydrothermal vein systems. The data above are in good agreement with the experience from the Pannonian Basin and from the Colorado River delta which show that smectite practically disappears from the mixed-layer structure ($S = 0\%$) above 230°C (Fig. 6).

The variation of R_m with $S\%$ in the study area can be compared with that observed in some sedimentary basins where similar studies have been carried out (Hámor-Vidó & Viczián 1993) (see Fig. 7). The trend line inferred from Fig. 5 for the southern Benue Trough is shown on this figure. The trend line indicates the difference between diagenetic grades in the marginal areas ($S \approx 25\%$) and the axial zone ($S = 0\%$) although the exact slope of the line is rather uncertain due to the scatter of the data points near $S \approx 25\%$ and the lack of additional experimental points in the range $S = 0$ to 25% . This approximate trend line fits well within the accuracy limits with other lines from different sedimentary basins where diagenesis is the result of normal subsidence and burial e.g. the deep Neogene depressions of the Pannonian Basin, the Transcarpathian Basin (Franců et al. 1990) and the Carboniferous of Silesia

(Środoń 1979). Short lived thermal effects (e.g. in Kyushu, Japan, in Miki et al. 1991) would cause another type of trend in the interval of $R_m < 2.1\%$ as shown by the trend line VIII in Fig. 7.

Maturity levels and petroleum source bed potentials

Previous work has shown the Coniacian Awgu Shales (upper part of the Nkalagu Formation) and the Late Campanian Nkporo Shale (lateral equivalence of the Enugu Shales) to have proven potentials as petroleum source beds in the southern Benue Trough (Petters & Ekweozor 1982; Ekweozor & Gormly 1983; Unomah & Ekweozor 1993). This interpretation is based on the concentration, type and thermal maturity of organic matter in these shales.

Organic petrographic studies of shales from the various formations in the study area were reported in Akande et al. (1992). These results suggest that the sampled Late Campanian shale beds (sample IH in this study) were affected by a normal burial diagenesis and their maturity reached the beginning of the oil window at the present outcrop level in the Ihe and Leru areas. These would have generated some unknown quantity of liquid petroleum and bitumens into shallow reservoirs. Shales of the Turonian Eze-Aku Formation (samples EZ) are mature with the prevalence of more gas prone organic constituents. The Albian Asu River Group Shales (samples AM, ISH and ENY) were affected by localised hydrothermal effects apart from normal burial. This has contributed to their overmaturity beyond the wet and dry gas generation stages probably since the end of the Santonian tectonic event.

Conclusions

1 - This study of the diagenetic transformation of Cretaceous shales in the southern Benue Trough shows that the axial zone of the Trough consisting of the Albian Asu River Shales have undergone a complex diagenetic history involving a normal regional burial superposed by tectonism and local heating effects around hydrothermal centres at Ishiagu, Enyigba and Ameri. IC values here are between $0.4-0.9 \text{ } ^\circ 2\Theta$, $S = 0\%$ and reflectance of vitrinite R_m ranges over broad limits between 2.46 to 4.30%. This suggests strong diagenetic to anchizonal metamorphic transformations.

2 - The Eze-Aku, Enugu/Nkporo Shales in the marginal areas were affected only by normal burial diagenesis resulting from subsidence. In this group, the IC values are between 1.4 and $1.6 \text{ } ^\circ 2\Theta$, $S = 20$ to 30% and R_m values vary from ca. 0.7 to 0.9%.

3 - Paleotemperature estimates were up to 230°C in the axial zone and about 100 to 140°C in the adjacent areas and the sampled locations.

4 - Albian shales in the axial part of the southern Benue Trough are generally overmature and consist of gas prone organic constituents. On the basis of their thermal maturity, we suggest that the Turonian to Campanian Shale units in the study area have undergone only a normal burial diagenesis within the oil generation zone. Given the right types of organic constituents, they could have generated some unknown quantity of hydrocarbons since attaining their present maturity levels.

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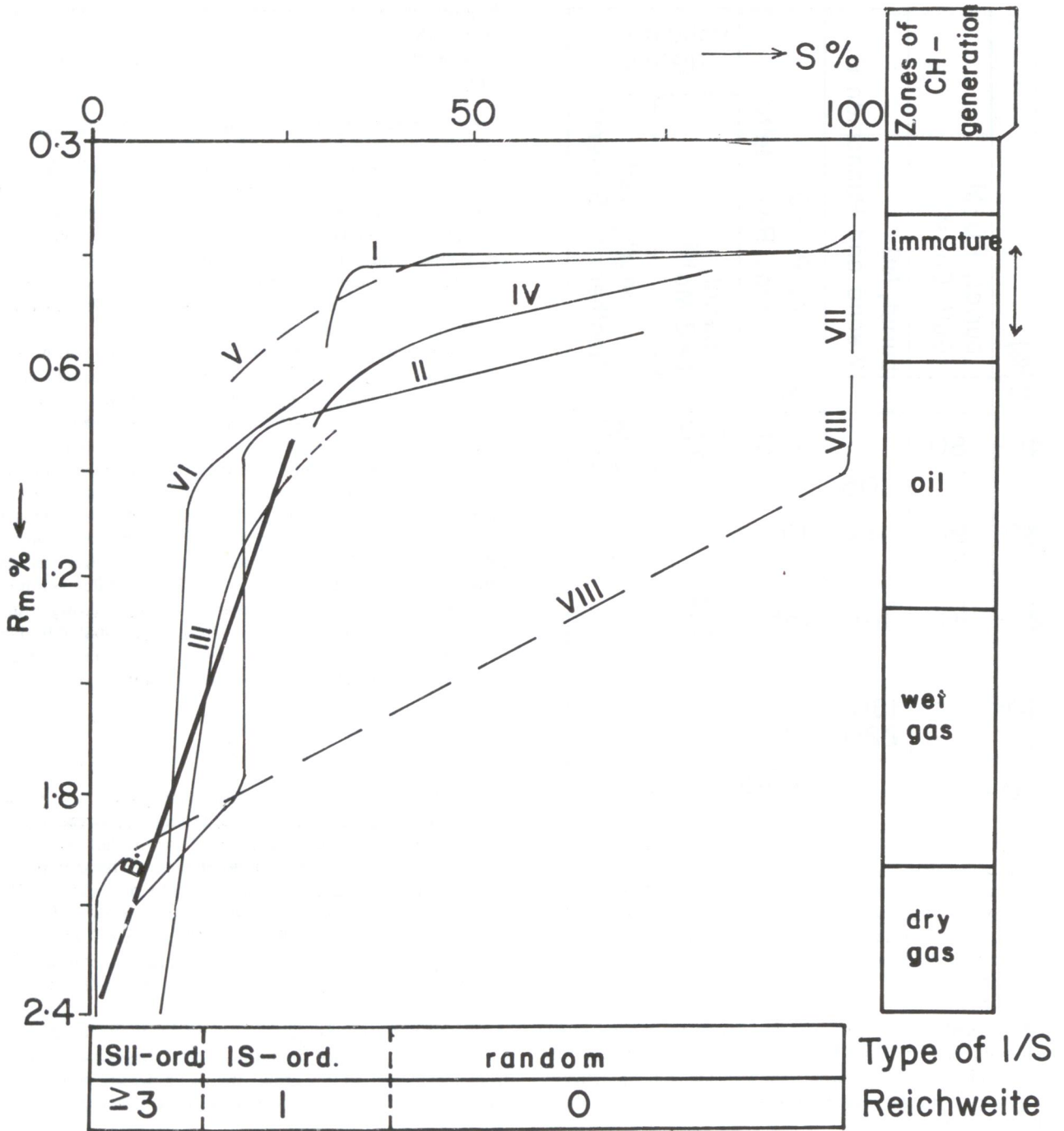


Fig. 7. Interrelations of R_m and S in various sedimentary basins. I: Pannonian Basin (thickness < ca. 2.5 km), II: Pannonian Basin, depressions (ca. 2.5 to 6 km thickness), I and II: data from Hámor-Vidó & Viczián (1993), III: Transcarpathian Basin, Slovakia, IV: Vienna Basin, Slovakia, III and IV: data from Francú et al. (1990), I-IV are Neogene successions. V: Tertiary, borehole Karlsefni H-13 off the coast of Labrador (Kübler 1984), VI: Carboniferous of Silesia, Poland (Środoń 1979), VII: Pannonian Basin, areas of extreme high heat flow (Hámor-Vidó & Viczián 1993), VIII: Tertiary of Kyushu, Japan (Miki et al. 1991). Double arrow on the right side of the diagram shows R_m interval of transformation of illite/smectites according to Kübler (1984, Figs. 5, 6). Trend line for the southern Benue Trough (line B.) is taken from Fig. 5 of the present study. R_m data are compared with the zones of CH generation (Tissot & Welte 1978). S data are compared with type of ordering of the illite/smectite interstratification (Reynolds & Hower 1970).

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References

- Agagu O.K. & Adighije C., 1983: Tectonic and sedimentation framework of the Lower Benue Trough, south east Nigeria. *J. of Earth Sci.*, 1, 267-274.

- Akande S.O., Hoffknecht A. & Erdtmann B.D., 1992: Environment of ore formation and anchizonal metamorphism in Pb-Zn-fluorite-barite deposits of the Benue Trough, Nigeria. *Geol. en Mijnbouw*, 71, 131-144.
- Árkai P., 1987: New data on the petrogenesis of metamorphic rocks along the Balaton Lineament, Transdanubia, W. Hungary. *Acta Geol. Hung.*, 30, 3-4, 319-338.
- Eberl D.D., 1993: Three zones for illite formation during burial diagenesis and metamorphism. *Clays and Clay Miner.*, 41, 1, 26-37.
- Ekweozor C.M. & Gormly J.R., 1983: Petroleum geochemistry of Late Cretaceous and Early Tertiary shales penetrated by the Akukwa-2 well in the Anambra Basin, southern Nigeria. *J. Petrol. Geol.*, 6, 2, 207-216.
- Francú J., Müller P., Šucha V. & Zatkalíková V., 1990: Organic matter and clay minerals as indicators of thermal history in the Transcarpathian depression (East Slovakian Neogene Basin) and the Vienna Basin. *Geol. Carpathica*, 41, 5, 535-546.
- Hámmor-Vidó M. & Viczián I., 1993: Vitrinite reflectance and smectite content of mixed-layer illite/smectites in Neogene sequences of the Pannonian Basin, Hungary. *Acta Geol. Hung.*, 36, 2, 197-209.
- Hower J., Eslinger E.V., Hower M.E. & Perry E.A., 1976: Mechanism of burial metamorphism of argillaceous sediment: 1. Mineralogical and chemical evidence. *Geol. Soc. Amer. Bull.*, 87, 5, 725-737.
- Jennings S. & Thompson G.R., 1986: Diagenesis of Plio-Pleistocene sediments of the Colorado River delta, Southern California. *J. Sed. Petrology*, 56, 1, 89-98.
- Kisch H.J., 1987: Correlation between indicators of very low-grade metamorphism. In: Frey M. (Ed.): *Low temperature metamorphism*. Blackie, Glasgow and London, 227-346.
- Kübler B., 1964: Les argiles, indicateurs de métamorphisme. *Rev. Inst. Franç. Pétrole*, 19, 10, 1093-1112.
- Kübler B., 1984: Les indicateurs des transformations physiques et chimiques dans la diagenèse. Températures et calorimétrie. In: Lagache M. (Ed.): *Thermométrie et barométrie géologiques vol. 2, Ch. 14. Soc. Franç. Min. Crist.*, Paris.
- Kübler B., 1990: "Cristallinité" de l'illite et mixed-layers: brève révision. *Schweiz. Mineral. Petrogr. Mitt.*, 70, 89-93.
- Lippmann F., 1982: The thermodynamic status of clay minerals. In: van Olphen H., Veniale F. (Eds.): *Intern. Clay Conf., 7th, Bologna and Pavia, 1981. Developments in Sedimentology 35, Elsevier*, Amsterdam etc., 475-485.
- Maxwell D.T. & Hower J., 1967: High-grade diagenesis and low-grade metamorphism of illite in the Precambrian Belt series. *Amer. Mineralogist*, 52, 5-6, 843-857.
- Miki T., Nakamuta Y. & Aizawa J., 1991: Relationships between authigenic mineral transformation and vitrinite reflectance during diagenesis: an example from the Tertiary of northern Kyushu, Japan. *Clay Miner.*, 26, 2, 179-187.
- Petruk W., 1964: Determination of the heavy atom content in chlorite by means of the X-ray diffractometer. *Amer. Mineralogist*, 49, 1-2, 61-71.
- Petters S.W., 1977: Mid-Cretaceous paleoenvironments and biostratigraphy of the Benue Trough, Nigeria. *Bull. Geol. Soc. America*, 89, 151-154.
- Petters S.W. & Ekweozor C.M., 1982: Petroleum geology of Benue Trough and south-eastern Chad Basin, Nigeria. *Amer. Assoc. Petrol. Geologists Bull.*, 66, 1141-1149.
- Pollastro R.M., 1990: Clays. In: Magoon L.B. (Ed.): *The petroleum system - status of research and methods, 1990. US Geol. Surv. Bull.*, 1912, 28-31.
- Reynolds R.C. & Hower J., 1970: The nature of interlayering in mixed-layer illite-montmorillonites. *Clays and Clay Miner.*, 18, 25-36.
- Rischák G. & Viczián I., 1974: Mineralogical factors determining the intensity of basal reflections of clay minerals. *MÁFI Évi Jel. (Annual Rept. of Hung. Geol. Inst.) 1972*, 229-256 (in Hungarian, English abstract).
- Šrodoň J., 1979: Correlation between coal and clay diagenesis in the Carboniferous of the Upper Silesian Coal Basin. In: Mortland M.M. & Farmer V.C. (Eds.): *Intern. Clay Conf., 6th, Oxford, 1978. Developments in Sedimentology 27, Elsevier*, Amsterdam etc., 251-260.
- Šrodoň J., 1980: Precise identification of illite/smectite interstratifications by X-ray powder diffraction. *Clays and Clay Miner.*, 28, 6, 401-411.
- Šucha V., Kraus I., Gerthofferová H., Peteš J. & Sereková M., 1993: Smectite to illite conversion in bentonites and shales of the East Slovak Basin. *Clay Miner.*, 28, 243-253.
- Tissot B.P. & Welte D.H., 1978: Petroleum formation and occurrence. *Springer*, Berlin, etc.
- Unomah G.I. & Ekweozor C.M., 1993: Petroleum source rock assessment of the Campanian Nkporo Shale, Lower Benue Trough, Nigeria. *Nigerian Ass. Petroleum Explorationists Bull.*, 8, 2, 172-186.
- Viczián I., 1967: Erfahrungen mit der Anwendung der Röntgendiffraktometrie zur quantitativen Bestimmung sedimentärer Mineralien. *MÁFI Évi Jel. (Annual Rept. of the Hung. Geol. Inst.) 1965*, 567-576 (in Hungarian, German and Russian abstracts).
- Viczián I., 1985: Diagenetic transformation of mixed-layer illite/smectites in deep zones of the Pannonian Basin (Hungary). *5th Meeting of the European Clay Groups, Prague, Czechoslovakia, 1983, Univerzita Karlova, Praha*, 135-140.
- Viczián I., 1994: Smectite-illite geothermometry. *Földt. Közl.*, 124, 3, 367-379 (in Hungarian, English abstract).