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## Palynological and Organic-Petrographic Data on Very Low- and Low-grade Metamorphic Rocks in the Slavonian Mountains (Northern Croatia)

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**Key words:** Palynomorphs, Macerals, Thermal maturity, Very low- and low-grade metamorphic rocks, Stratigraphy, Palaeoenvironments, Slavonian Mts., Croatia

### Abstract

The palynological data suggest a Late Silurian to ?Early Carboniferous sedimentation age for the weakly metamorphosed rocks from the Mts. Psunj, Papuk and Krndija (Slavonija, North Croatia). Palynofacies and organo-petrographic data indicate that the protolithic sedimentary rocks were deposited in comparatively calm, anoxic to suboxic depositional environments. The vitrinite reflectance data for the same samples indicate the coal rank of meta-anthracite or anchimetamorphic zone grading into the coal rank of meta-anthracite/semigraphite or anchimetamorphic/epimetamorphic zone.

### 1. INTRODUCTION

The age of some of the parts of the Slavonian Mountain metamorphic complexes, the rocks of which stretch from the westernmost parts of Mt. Psunj towards Mt. Papuk and to the easternmost parts of Mt. Krndija, has not been to date reliably defined. This also partly holds for the weakly metamorphosed rocks which are widespread in this area (Fig. 1). The aim of this paper is to present palynological and organo-petrographic data obtained on very low- and low-grade metamorphic rocks, which were sampled along a cross-section located from the westernmost to the easternmost parts of the metamorphic complexes of the Slavonian Mountains (Fig. 1), in order to contribute to the knowledge of the stratigraphic position of these metamorphic rocks.

### 2. LITERATURE REVIEW

Numerous papers have been published on the age of the crystalline rocks from different parts of the Slavonian Mountains. In the earliest papers, the opinion prevailed that highly metamorphosed rocks belonged to the Precambrian and weakly metamorphosed rocks to the Palaeozoic and Carboniferous respectively (GOR-

JANOVIĆ-KRAMBERGER, 1897; KOCH, 1919; POLJAK, 1934). Later, POLJAK (1952), identified the alleged Silurian graptolites in slates of the weakly metamorphosed, i.e. Radlovac complex from the northeastern parts of Mt. Papuk. ŠIKIĆ & BRKIĆ (1975) discounted Poljak's graptolite determinations when they ascertained that the graptolites were, in fact, inorganic "load casts". In slates from the outcrop at Mt. Papuk where Poljak collected his graptolites, a Carboniferous (Westphalian) macroflora was reported by BRKIĆ et al. (1974). On the basis of this palaeontological data and field relations, JAMIČIĆ (1983) concluded that the lowermost parts of the weakly metamorphosed Radlovac complex are Late Devonian and the highest parts of the complex are Late Permian in age.

The first radiometric data on associated highly metamorphosed rocks from the Slavonian Mountains were Rb-Sr measurements of 279 to 285 Ma by DELEON (1969) on migmatites from Mt. Papuk. On the basis of geological, petrological and structural data, JAMIČIĆ (1983, 1988) recognized three units in this area: (1) the Balkalian Psunj (or Psunj-Kutjevo) complex which consists mostly of gneisses, mica schists, amphibolites, granites, marbles, greenschists and phyllites; (2) the Caledonian Papuk (or Papuk-Jankovac) migmatitic complex with granitoids, and (3) the Hercynian weakly metamorphosed Radlovac complex, which is mostly composed of slates, phyllites, and metasandstones intruded by metabasic rocks. Recently, PAMIĆ et al. (1988) published K-Ar and Rb-Sr ages obtained on about 50 monomineralic concentrates and whole-rock samples of highly crystalline rocks from the Slavonian Mountains. Measurements carried out on migmatites and associated S-type granitoids yielded K-Ar ages of 336 to 272 Ma, and two Sr-isochron ages of 314 and 317 Ma. Consequently, there is little doubt that the migmatitic-granitic complex is Hercynian in age. By contrast, K-Ar measurements on hornblende and biotite concentrates from amphibolites and gneisses of the progressive metamorphic (Psunj or Psunj-Kutjevo) complex yielded three groups of ages ranging from 262 to 262, 376 to 352 and 658 to 422 Ma. Based in part on correlation of the age of the granitoids and migmatites, the preferred age of the main phase of metamorphism is given by the hornblende K-Ar ages ranging from 353 to 376 Ma.

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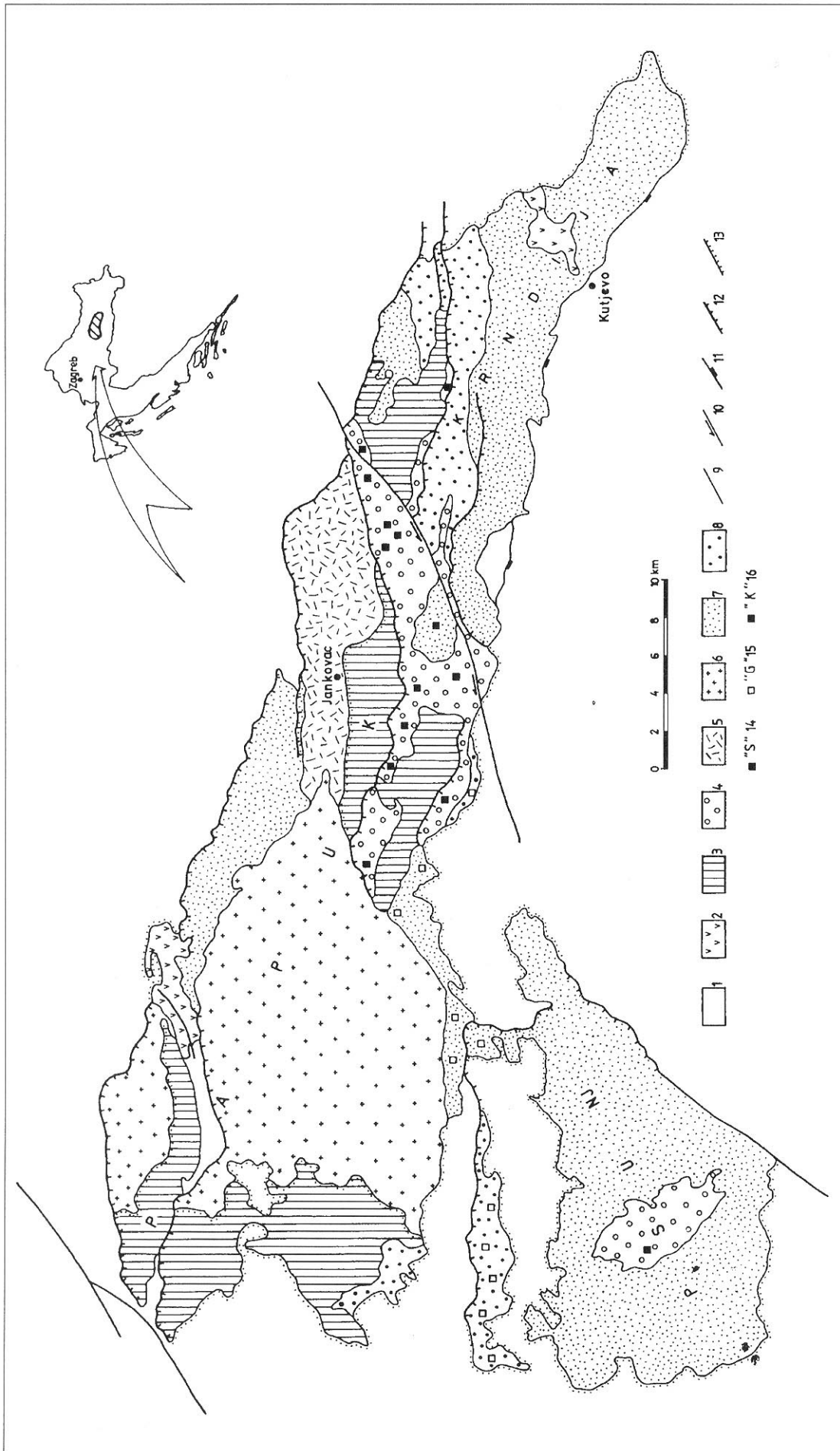


Fig. 1. Geological map of the Mts. Psunj, Papuk and Krndija (simplified version of the map presented in PAMIĆ & LANPHERE, 1991a). Legend: 1) Tertiary and Quaternary sediments of the Pannonian basin; 2) Tertiary volcanic rocks; 3) Mesozoic, mostly Triassic rocks; 4) Hercynian weakly metamorphosed complex with metabasic igneous rocks; 5) Hercynian migmatites; 6) S-type granitoids; 7) Progressively metamorphosed complex, mostly amphibolite facies, in places with I-type granites; 8) Progressively metamorphosed complex, mostly greenschist facies; 9) Contact line; 10) Horizontal fault; 11) Normal fault; 12) Reverse fault; 13) Unconformity; 14) Palynological samples from the weakly metamorphosed rocks; 15) Palynological samples from the lowermost parts of the progressive metamorphic complex (samples without chloritoids); 16) Palynological samples from the lowermost parts of the progressive metamorphic complex (samples with chloritoids).

### 3. BASIC GEOLOGICAL DATA

In the area of the Slavonian Mountains and the surrounding basement of the Pannonian Basin three main magmatic-metamorphic complexes can be distinguished: (1) the progressive metamorphic complex with I-type granites which was metamorphosed mostly during the Hercynian deformational event under P-T conditions of amphibolite and greenschist facies; (2) the Hercynian migmatitic-granitic complex which evolved continuously from the progressive metamorphic complex in the area of increased geothermal gradients, and (3) the weakly metamorphosed (Radlovac) complex which was metamorphosed under very low- and low-grade conditions (PAMIĆ & LANPHERE, 1991b).

Accordingly, weakly metamorphosed rocks sampled for palynological research come from the Radlovac complex ("S" samples) and from the lowermost parts of the progressive metamorphic complex (samples "G" without chloritoids and samples "K" with chloritoids). These three groups of samples are presented by different symbols on the geological map (Fig. 1).

The weakly metamorphosed rocks from both complexes are mostly represented by slates and phyllites interlayered with schistose metasandstones grading into quartz-sericite schists. Petrogenically, weakly metamorphosed rocks from both complexes are complementary to each other because the maximum metamorphic grade of the Radlovac complex is the lowermost part of the greenschist facies, whereas the progressively metamorphosed complex starts with middle to upper parts of greenschist facies (PAMIĆ & LANPHERE, 1991b).

### 4. MATERIALS AND METHODS

Samples for palynological investigation were collected from the central parts of Mt. Psunj, the central and northwestern parts of Mt. Papuk and in the adjoining area of the Papuk and Krndija Mountains (Fig. 1).

Carbonates were dissolved in 15% HCl (24 hours), and the silicates in 40% HF (10 days). The obtained residuum was treated with concentrated HNO<sub>3</sub> (3 hours at 60° C) to aid oxidation and elimination of pyrite (sulphides). The organic residuum was separated by flotation in ZnCl<sub>2</sub> (s.g. »2.1 kg/l), and then separated using a 20 µm sieve. The palynomorphs were black and barely transparent, and required further treatment with SCHULZ solution (HNO<sub>3</sub> and KClO<sub>3</sub>). Palynological slides were mounted using glycerine-jelly.

Silicone polished block were also prepared from the organic residuum for organo-petrographic analysis and vitrinite reflectance.

### 5. PALYNOLOGICAL DATA

In several "S" samples from the Radlovac complex, rare, poorly preserved palynomorphs were found. The

sphaeromorphitic Acritarchs, such as *Protoleiosphaeridia*, and *Leiosphaeridia* predominate over extremely scarce sporomorphs such as cf. *Cyrtospora*, cf. *Ambitisporites*, cf. *Dyadospora* and fungal spores. A few chitinous forms (microforaminifera, tintinids and scolecodonts) were also found (Plate I). No acanthomorphitic Acritarchs were observed.

In only one "G" sample from rocks of the progressive metamorphic complex, scarce, poorly preserved *Protoleiosphaeridia* were observed. In the "K" chloritoid bearing samples no palynomorphs were observed.

High thermal degradation of the collected palynomorphs does not allow more precise taxonomical determination. Potentially chronostratigraphically important sporomorphs are approximately determined at the generic level, whereas somewhat better preserved phytoplanktonic forms have greater stratigraphic range. Nevertheless, a Late Silurian to ?Early Carboniferous age can be suggested for these samples. *Ambitisporites* characterize the Silurian (STREEL & TRAVERSE, 1978; STROTHER & TRAVERSE, 1979) and extend, according to TRAVERSE (1988), into the Early Devonian. *Dyadospora* was recorded from the Silurian deposits (STROTHER & TRAVERSE, 1979; BARRON, 1989). *Cyrtospora* is believed to range from the Late Devonian to the Early Carboniferous (VAN DER ZWAN, 1979; VAN DER ZWAN & WALTON, 1981; TRAVERSE, 1988). Each of the mentioned sporomorphs occurs in different slides of the "S" samples (Plate I), suggesting the possibility that this sporomorph assemblage may represent, in part, sediment reworking.

Despite the uncertain stratigraphical attribution of these samples, the vitrinite-bearing "S", "G" and some of the "K" samples provide important information on the lowermost age of the strata. The first occurrences of the vitrinite clasts (=wood fragments) are closely related to the first appearance of the first vascular plants in the Late Silurian (TRAVERSE, 1988), indicating that these vitrinite-bearing samples are not older than the Late Silurian.

Due to the approximate taxonomic determinations, the uppermost, i.e. Lower Carboniferous age of these sample has to be treated extremely tentatively.

Among the Carboniferous macroflora of the Radlovac complex, BRKIĆ et al. (1974) reported poorly preserved grains of *Cordaitina*-type sporomorphs which are not known from sediments older than the Carboniferous. This horizon was also palynologically checked in our investigation. The organic matter of this sample is composed of well-rounded vitrinite/inertinite clasts, indicative of reworking, but no isochronous (Carboniferous) palynomorphs were observed. This could be explained by the extremely hostile environments for the preservation of the palynomorphs during synsedimentary reworking in an anoxic environment. From this point of view it is possible that the Silurian-Devonian sporomorphs identified in this sample were reworked into the Carboniferous sediments.

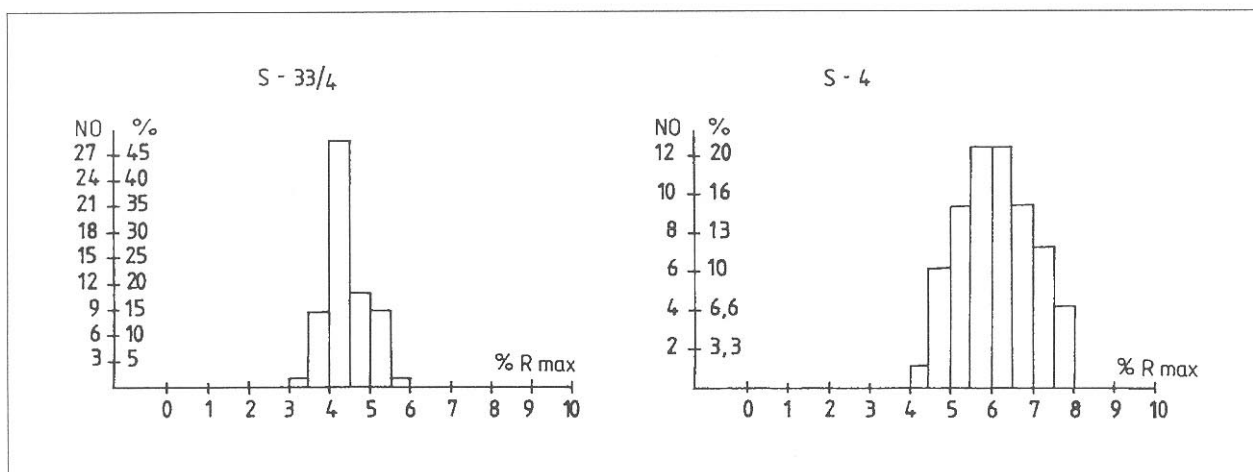


Fig. 2. The values of vitrinite reflectance indicate coal rank of meta-anthracite or anchimetamorphic zone (S - 33/4) and coal rank of meta-anthracite/semigraphite or anchimetamorphic/epimetamorphic zone (S - 4).

## 6. PALYNOFACIES AND ORGANO-PETROGRAPHIC ANALYSIS

In the "S" samples the organic matter is composed predominantly of micrinite (60-80%) and angular to subangular clasts of vitrinite/inertinite (20-40%). Only one sample contains organic matter of pure vitrinite/inertinite represented by well-rounded clasts. In the "G" and "K" samples the amount of organic matter is small but are characteristically very similar to those in "S" samples.

## 7. PALAEOENVIRONMENT OF THE PROTOLITHIC ROCKS

The predominance of the phytoplanktonic forms over the scarce sporomorphs and the composition of the organic matter in which micrinite (= thermally changed amorphous liptinite) predominates over poorly rounded vitrinite/inertinite, indicates a comparatively calm, anoxic to suboxic marine depositional environment with some terrestrial influence. It is interesting that no acantomorphic Acritarchs, which are otherwise more or less common components of Palaeozoic marine paly-nofacies, have been observed. The lack of this type of Acritarchs could be interpreted as selective distribution of the palynomorphs, though the possibility of facies-control of the palynofloral assemblage can not be excluded. Only one sample (showing a Carboniferous macroflora) with pure well-rounded vitrinite/inertinite clasts indicates a fluvial or near-shore oxic depositional environment.

Although the organic matter of the "G" samples is similar to that in the "S" samples, data obtained from "G" and "K" samples, generally, are insufficient for palaeoenvironmental conclusions.

## 8. THERMAL MATURITY

### 8.1. SPORE COLORATION

It is a well known fact that the sporonine progressively changes during the irreversible processes of catagenesis by increasing temperature. Colour changes in palynomorphs depend on their structure and thickness of the exine, palynomorph type and the duration of the process. Palynomorphs of phytoplanktonic origin respond more slowly to thermal changes than the sporomorphs. Colour changes enable the determination of palaeotemperatures up to around 200°C (DORNING, 1985; BARRON, 1989). Thus, the black colour of the analysed palynomorphs indicates temperatures in excess of 200°C.

### 8.2. VITRINITE REFLECTANCE

The method of vitrinite reflectance used on organic matter thermally altered between 200°C and 400°C gives more precise values. One "S" sample (Fig. 2) shows  $\%R_{max} = 4.39 \pm 0.51$  indicating the coal-rank of meta-anthracite or the anchimetamorphic zone (300-350°C). The second "S" sample shows higher values of  $\%R_{max} = 6.10 \pm 0.33$ , indicating the coal rank of the meta-anthracite/semigraphite or anchimetamorphic/epimetamorphic zone (350°C) (ARKAI, 1983, 1987; PAMIĆ et al., 1992).

## 9. DISCUSSION

The palynological data obtained from very low- and low-grade metamorphic rocks of the Slavonian Mts. have to be considered as preliminary. This is mainly due to (1) the rather small number of analysed samples covering a comparatively big area, (2) scarce and poorly preserved palynomorphs, (3) the limiting factor in the investigation of thermally highly altered palynomorphs, is the lack of a method for simultaneously



analysing palynomorphs in reflected and transmitted light, and (4) the lack of an inadequate preparation method (REITZ, pers. comm.). Nevertheless, on the basis of the palynological data obtained it is inferred that the sedimentation age of the "S" samples occurred within the range Late Silurian-?Early Carboniferous. The vitrinite reflectance and spore coloration indices suggest that these samples were affected by very low-grade metamorphism.

Our data can be correlated with some adjacent semi-metamorphic complexes. Similar correlatives of the very low- and low-grade metamorphic rocks occur in Western Carpathia, the "Gelinca Terrane" (southern Gemic zone), composed of "flyschoid" sediments interstratified with volcanic rocks. The microfloristic data indicate a sedimentation age from the Late Cambrian to the Devonian. The whole complex was metamorphosed to the lower parts of the greenschist facies (VOZAROVA & VOZAR, 1992).

In the Hungarian Transdanubian Midmountains sedimentation ages of the very low-grade metamorphic rocks range from the Lower Ordovician to the Lower Carboniferous. The whole complex is composed of metasilstones and metasandstones intercalated with metapelites. The Lower Ordovician and Lower Silurian age of the lower part of this complex is supported by palynological data (ARKAI & LELKES-FELVARI, 1992).

The largest part of the Northern Graywacke Zone ("Upper Austroalpine" in the Eastern Alps) is composed mostly of pelitic, low-grade metamorphic rocks. The microfloristic analysis revealed the Lower Ordovician age of these metapelitic rocks (REITZ & HÖLL, 1991).

Within the metasedimentary frame of the gabbroamphibolite complex in south Germany near the Bohemian boundary, quartz micaschists of the lowest grade of metamorphism occur. These mica schists were palynologically investigated and they have yielded a sporomorph assemblage indicating a Middle to Upper Silurian age (REITZ, 1992). It should be emphasized that this gabbroamphibolite complex was previously considered as Precambrian by German and Bohemian geologists.

Palynostratigraphical correlation between the weakly metamorphosed Radlovac complex ("S" samples) and the lowermost parts of the progressive metamorphic complex (samples without chloritoids and samples "K" with chloritoids) is questionable. Although petrogenetically justified, palynostratigraphical correlation between these two complexes was not possible, primarily due to the extreme palynomorph scarcity of greater stratigraphical range in the "G" samples and a total lack of palynomorphs in the "K" samples. However, it must not be overlooked that the oxidation, which was performed in order to improve the transparency of the thermally high altered palynomorphs of the "G" and "K" samples, did not give any expected results, meaning that the palynological slides are per-

haps not barren (this is also true for the heavy liquid separation, during which sporomorphs could have been lost). Such thermally highly altered palynomorphs often appear in palynological slides as totally black, opaque clasts without any recognizable morphology and structure. However, the simultaneous observation in reflected and transmitted light enables the recognition of the morphological features as shown in numerous publications of Reitz (for references see REITZ, 1992). For that reason, the future palynological investigation must enhance the use of the mentioned method.

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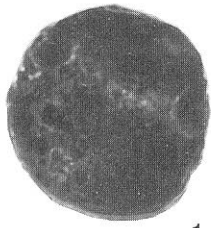
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#### PLATE I

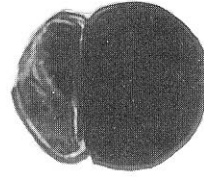
1. cf. *Ambitisporites*; proximal side. S-4/2, 600X.
2. cf. *Cyrtospora*; proximal side. S-9/1, 600X.
3. cf. *Dyadospora*; diad. S-14/2, 600X.
4. Fungi, *Sporites* gen. et sp. indet. S-19/2, 600X.
- 5, 6. *Protoleiosphaeridia* sp.; 5. S-10/3, 6. G-3/1, 600X.
7. *Leiosphaeridia* sp.; S-24/1, 600X.
8. Cluster of *Leiosphaeridia* sp.; S-33/3, 400X.
9. Microforaminifera S-9/2, 600X.
- 10, 11. Chitinous lorica of tinitinid; S-33/4, 600X.
12. Scolecodonta; S-33/1, 600X.
13. Vitrinite, well structured fragment; S-33/1, 600X.



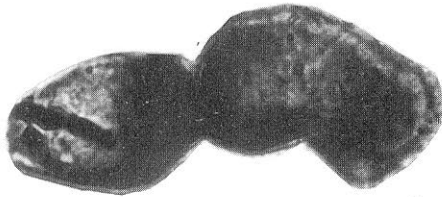
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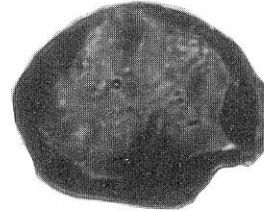
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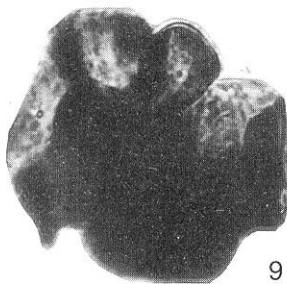
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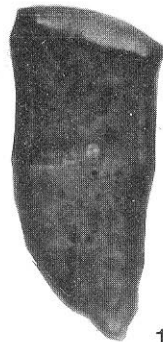
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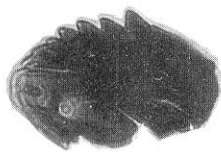
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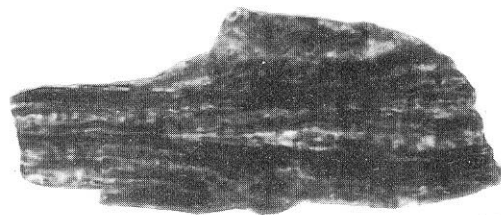
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