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GEOLOGICAL IMPORTANCE OF SOME CHEMICAL FEATURES OF COALS IN HUNGARY

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ABSTRACT

Geochemical characteristics of sedimentary rocks, including coals and especially those of their ashes and detrital rocks, respectively, are primarily determined by geological setting of the denudation area. Significant influencing factors are supposedly the weathering factors, especially the climate. It seems to be possible to approach the question of climate by analysing the modulus of coal ashes.

INTRODUCTION

During the investigation of distribution of major elements of variegated clays from the Transdanubian Central Mountains (JÁMBOR *et al.*, 1968) it was evidenced that the modulus of the clayey rocks shows comparatively significant variation in different eras of the Earth's history. As far as both pelitic and variegated clayey rocks were concerned the significant role of climatic differences was thought of as possible cause of differences.

The collection and publication of data concerning coal beds being mined regularly in Hungary (KOVATSITS and WOLF, 1980) provided the possibility for further consideration. Data in this publication cited contain three-month averages of the daily collected quality control data for the different mining products. From this publication mainly data, concerning ash content were used furthermore the geological relations of total water and organic carbon content of coals were also studied. As a first step of data processing the analytical data from the 25 plants operating in the country, arranged into 7 groups according to geological age (Table 1) were averaged (Table 2). For better understanding both Table 1 and Table 2 include these data.

DISCUSSION

Modulus

The modulus (Al_2O_3/SiO_2) is a value used mainly in the evaluation of bauxites and it is dependent on the quantitative and qualitative relation of allitic and siallitic components in bauxites. In clayey rocks containing allitic components this value is

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TABLE 1

Geological age and some characteristics of mined coal deposits in Hungary

Occurrence and age	Classification	Country rocks
1. Mecsek (Liassic)	medium to high volatile bituminous	argillite, aleurolite and sandstone
2. Ajka (Upper Cretaceous)	high volatile bituminous C	clayey marl, marl
3. North Transdanubian (Eocene)	subbituminous A	clay, aleuritic limestone
4. Tokod (Oligocene)	subbituminous C	clay, aleurite
5. Nógrád and Borsod (Lower Miocene)	lignite A	clay, aleurite
6. Várpalota (Middle Miocene)	lignite B	clay, oil shale
7. Mátraalja (Pliocene)	lignite B	clay, aleurite, sand

over 1.0, in average clayey rocks from 0.2 to 0.5 but can reach the value of 1.0, which is over the value for pure kaolinite (0.85). It means that the modulus is connected with the chemical composition of claystones as well and, being related to the progress of diagenesis, it is significantly influenced by the intensity of burial, namely diagenesis tends to continually reduce the modulus in an extent unknown at present, since in great depth of 2000 to 3000 meters rocks contain only illites and chlorites of hardly known chemical composition, the modulus of which is lower than that of kaolinite. Naturally, in determining of modulus of a claystone besides clay minerals other mineral components, first of all the quantity of quartz and feldspar grains in the silty fraction are also important. Their ratio, however, is influenced by the climate just like that of clay minerals. Warmer and more humid climate results in the increase of the modulus.

It is evident that the modulus of claystones can not be considered as a measure for the definition of warm and humid climates but it may give a good point of orientation besides palaeontological data.

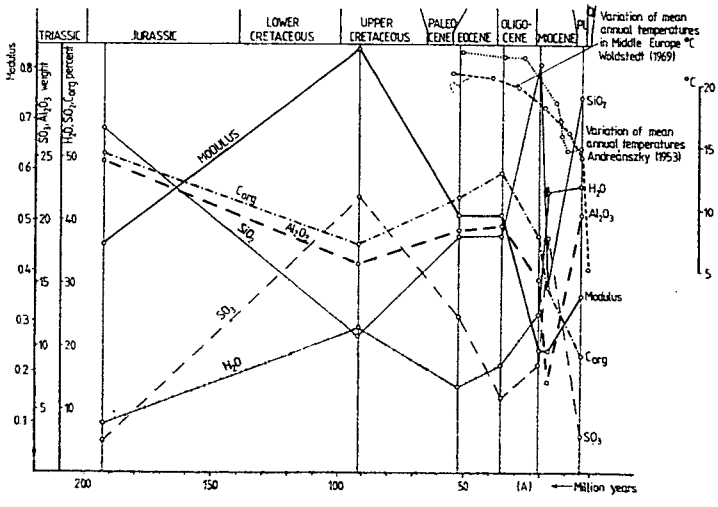

Fig. 1. Variation of averages of some chemical components in coals mined in Hungary

TABLE 2

Some components (in average) of Hungarian coals and their ashes

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	Total H_2O	Modulus	K_2O/Na_2O	CaO/MgO	C_{org}
1. Liassic	54.5	24.7	10.6	1.7	1.1	3.1	0.4	2.5	7.8	0.45	7.8	1.5	50.1
2. Upper Cretaceous	19.7	16.5	8.9	28.5	3.4	0.3	0.3	21.7	22.8	0.84	1.0	8.4	36.0
3. Eocene	37.8	19.1	9.2	11.8	3.5	1.1	1.6	12.3	13.9	0.51	0.7	3.4	43.8
4. Oligocene	37.9	19.6	20.6	7.0	4.3	1.4	3.0	6.0	15.1	0.51	0.46	1.6	47.6
5. Lower Miocene	64.3	15.2	10.5	8.8	2.3	1.4	1.2	8.7	25.0	0.24	1.2	3.8	37.5
6. Middle Miocene	29.5	7.1	7.7	28.9	4.4	1.6	0.8	18.7	44.1	0.24	2.0	6.6	29.0
7. Pliocene	59.3	20.5	6.4	4.8	2.0	0.9	0.2	2.9	45.3	0.35	4.5	2.2	18.4

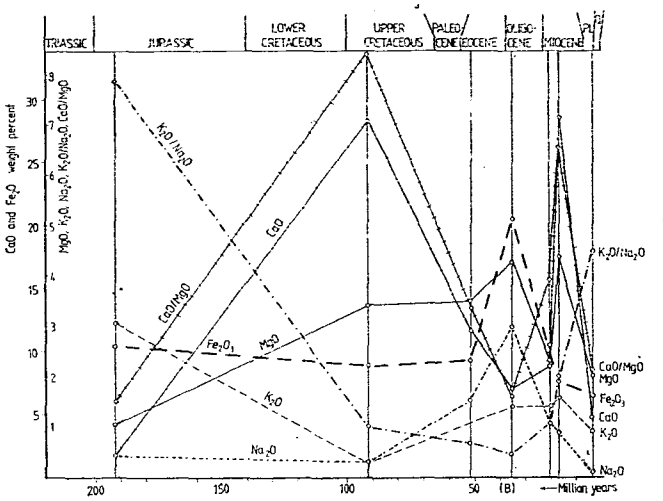

Fig. 2. Variation of averages of some chemical components in coals mined in Hungary

The modulus of Hungarian Upper Cretaceous coals is extremely high (0.84). It is in close concordance with the data from palynological analyses (GÓCZÁN, 1972). These show that climate in the Upper Cretaceous was tropical warm and humid but still insufficient for the formation of bauxites. The modulus is sharply decreasing from the Upper Cretaceous to the Badenian. The value for Liassic coal ashes is slightly lower than that for Eocene and Oligocene coals, whereas the modulus of Pliocene (Upper Pannonian) lignite is again higher. Decrease from the Upper Cretaceous to the Middle Miocene is interpreted as evidence of decrease in temperature. The modulus of Eocene and Oligocene coals is similar although in the Oligocene

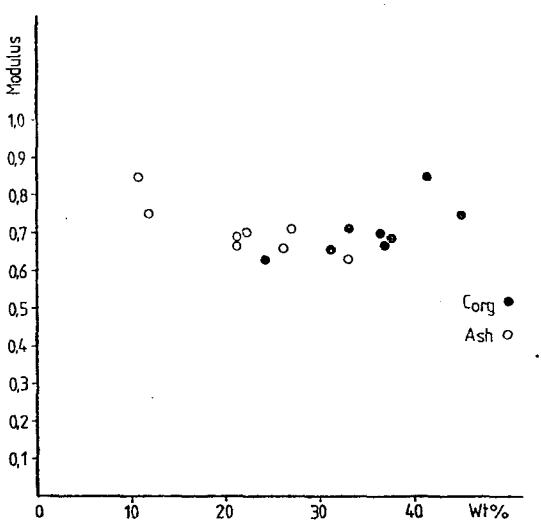

Fig. 3. Relation of modulus — ash and modulus — C_{org} in Upper Cretaceous coals

a cooler climate was unambigously evidenced. This similarity can be explained by the more humid climate in the Oligocene.

The decrease of modulus in the Liassic can be interpreted by a more disadvantageous climate on one hand and an advanced stage of the diagenesis on the other (Table 1). Increase of the modulus in the Pliocene can be regarded as sign of a lower degree of diagenesis as compared to that in the Miocene.

Chemical composition of ashes of the mining products depends on two components. These are on one hand inorganic components taken up from the soil by the plants and incorporated in their body and preserved during the process of coalification and embedding rocks inevitably entering the product during mining and processing as well as the nature of embedded rock fragments on the other.

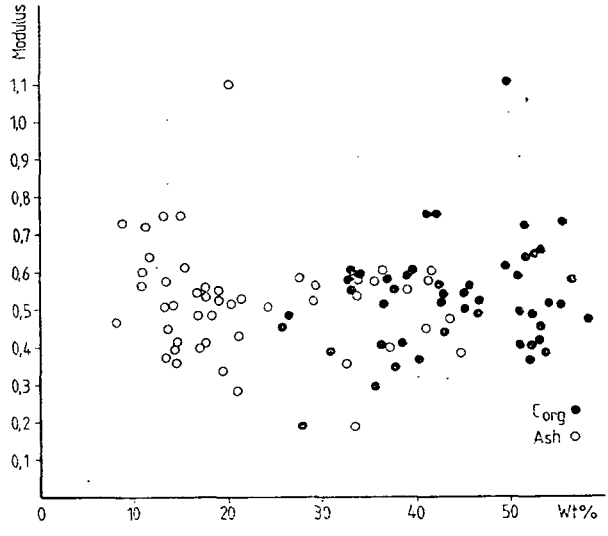

Fig. 4. Relation of modulus — ash and modulus — C_{org} in Eocene coals

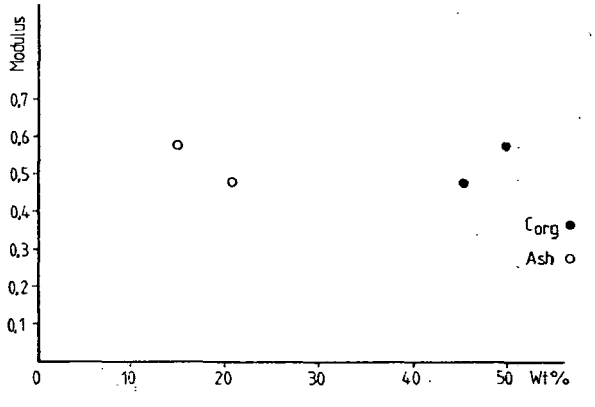

Fig. 5. Relation of modulus — ash and modulus — C_{org} in Oligocene coals

Using the possibility of analyses it was investigated whether the coal-rich products or products containing less coal have higher modulus and whether there is a difference between the modulus of coal ashes and that of barren rocks. *Figs 3* to *6* unambiguously show that finer coal products are cleaner. At the same time their modulus in Upper Cretaceous, Lower Eocene, Oligocene and Lower Miocene coals is in most cases higher. Thus the material taken up and incorporated by the plants evidences higher temperature, a more humid climate than do the bedrocks.

Naturally, SiO_2 and Al_2O_3 content of coal ashes unequivocally evidences the changes in the denudation area. From the Liassic to the Oligocene both factors are of similar quantity. In the Lower Miocene a large gap was formed between the two factors in favour of SiO_2. The situation remained the same in the course of the history of Earth in accordance with palaeogeographic facts.

Sulphur content

Sulphur content of the coal-ashes, as it is known since long time, depends on the abundance of carbonate rocks on the denudation area. This is unequivocally proved by the variation of sulphur content in Hungarian coals of different geological age. Liassic coals from the Mecsek Mountains contain the least sulphur. It proves that although a theoretical possibility of an outcrop of Middle Triassic carbonate com-

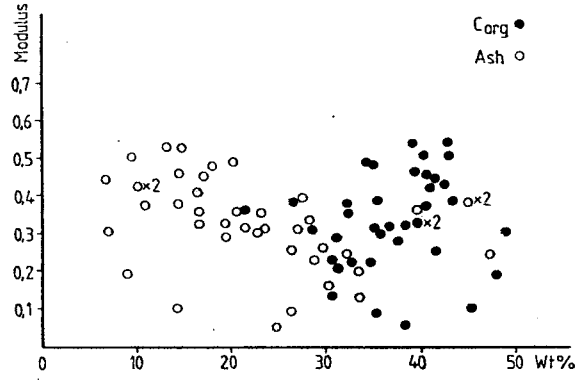

Fig. 6. Relation of modulus — ash and modulus — C_{org} in Lower Miocene coals

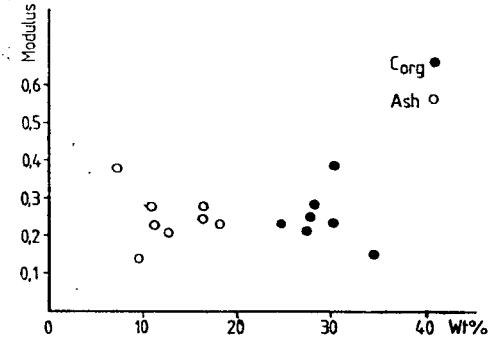

Fig. 7. Relation of modulus — ash and modulus — C_{org} in Middle Miocene coals

plexes existed in the SW foreland of the Mecsek Mountains (NAGY, 1962), the chemistry of coals indicates that the denudation area was absolutely free from carbonate rocks.

The high sulphur content of the Upper Cretaceous coal beds of Bakony, Ajka is in close relation with the well known palaeogeographical picture (HAAS, 1979), namely that carbonate rocks played an important or determinative role in the denudation area. Transgression over the Mesozoic limestone-dolomite mass of the Transdanubian Central Mountains is well perceivable even in the course of the transgression in the Lower Eocene. At the beginning of the Oligocene there are hardly any carbonate masses on the surface, most part of the Transdanubian Central Mountains became sedimentary area again. Here, sulphur content is again in concordance with the knowledge obtained in the course of the thorough complex analysis (KORPÁS, 1981). These facts are important, since part of the specialists repeatedly arrived at different conclusions from the present picture of a block mountain.

The medium sulphur content of the Upper Miocene coals of North Hungary and the high sulphur content of the Middle Miocene coal seams of Várpalota is also in close concordance with the palaeogeographical facts (HÁMOR, 1981). In the surroundings of the Lower Miocene beds there were only few carbonate rocks on the denudation area. On the contrary the Middle Miocene coal swamp of Várpalota was surrounded by hills consisting of carbonate rocks.

Sulphur content of the Pliocene coal beds verifies that the denudation area was almost totally free of carbonate rocks, a fact evident from the known palaeogeographical summaries (BALÁZS *et al.*, 1981).

With a little boldness, on the basis of the close agreement of data it can be declared that from the sulphur content of the coals numerical data can be obtained concerning the role of the carbonate rocks in the denudation area.

CaO content of coal ashes, naturally, varies in close connection with the carbonate surroundings, consequently, also with the sulphur content, although no mineralogical connection exists between them. Calcium appears mainly in form of carbonates and sulphur is mainly found as pyrite.

MgO content is higher in carstic coals than in coals of non-carstic origin but whilst CaO dominates in carstic ones it never does in the case of non-carstic coals (Liassic, Oligocene, Lower Miocene, Pliocene). CaO/MgO ratio is in accordance with this, although there is some discrepancy in the Lower Miocene deposits.

Other inert components

The amount of total iron as Fe_2O_3, K_2O and Na_2O can be interpreted only with greater difficulty. Iron content is relatively stable ranging between 6 and 10 per cent. Disregarding the iron content (21 per cent) in the Oligocene beds, it shows a slightly decreasing tendency in the course of the Earth's history and a relatively close correlation with the Na_2O content can be established.

The geohistorical background of both the correlation of the two components and the anomalous values in the Oligocene beds is not known at present.

Variation of K_2O content can also be contradictory interpreted. Although the highest value in Liassic formations is in close connection with the potassium abundance in the granitic denudation areas. Minimum in the Upper Cretaceous and increasing K_2O values in the Eocene and Oligocene show that this value is low in coals of carstic type but the relatively high values in the Middle Miocene and low values in Pliocene are contradictory.

High K_2O/Na_2O ratio in the majority of cases indicates beds of carstic type well, but this ratio is contradictory in the Oligocene and Middle Miocene beds.

Water and organic carbon

Comparison of water and organic carbon (C_{org}) content in coals untangles a close reverse correlation. Both factors depend on the extent of epigenesis, *i. e.* on the progress of the coalification. The high water and low C_{org} content of the Upper Cretaceous coals is noteworthy. It proves the low epigenetic stage of the Upper Cretaceous beds which is the result of the thin cover, formed over them in the Tertiary. Unlike the above cases this provides data for palaeogeographic investigation since it evidences that the Upper Cretaceous deposits of Ajka and Padragkút were only covered by thin sedimentary layers in the course of the Paleogene as well as the Neogene time. Higher grade of coalification of the coals from Eocene beds can be explained by the fact that coals of Dorog, Tatabánya and Felsőgalla are mined in greater depth. Lower coalification of coals from the mines in Oroszlány and Dudar-Balinka definitely indicates a thinner Oligocene/Neogene cover.

Higher coalification of Oligocene coals revealed in their C_{org} content as compared to Eocene ones is only apparent. Low C_{org} content of the Eocene coal products is the result of pollution occurring in the mining technology, a less frequent problem in Oligocene deposits, containing less barren embeddings.

Water content of coals continuously increased from 12 per cent in the Eocene to 46 per cent in the Pliocene. It seems that the total water content of the coals in this early stage of the epigenesis, namely in the upper gas zone, just like the free water content of clay minerals (HALMAI *et al.*, 1982) is a more sensible indicator of coalification than the vitrinite reflexion which changes here only between 0.2 and 0.5.

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