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THE OIL SHALE DEPOSIT OF VÁRPALOTA

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INTRODUCTION

After the discovery of the first Hungarian oil shale occurrence in the Balaton Highland near Pula village, in a maar-like tuff-ring, in 1973, we have paid much attention to Neogene sequences in confined or isolated basins. During mapping we have recorded alginite containing silty clay marls of Lower Pannonian age in shallow drill holes in the Kapolcs intramontane basin.

It was therefore necessary to carry out a thorough evaluation of the whole Transdanubian Mountain Range. The work was done by RAVASZ in 1977. In the course of these evaluations several samples were collected, one of which form an abandoned open pit at Bántapuszta near Várpalota by Cs. MUNTYÁN. According to thermal analysis this sample contained 47 weight % organic matter. This can be burnt and lit. While burning, it emanates a characteristic stearine odor. This finding has uprated the perspectivity of the Várpalota basin for finding oil-shales.

THE GEOLOGY OF THE VÁRPALOTA BASIN

The Várpalota basin lies in the southern foreland of the Bakony between Csór and Öskü *(Fig. 1)*. The basin shows a southward deepening. Its basement is composed of Silurian Carboniferous (?), Permian and Triassic rocks, getting gradually younger from the SE to the NW, being aligned in nearly SW-NE belts. The youngest member of the basement is the "Hauptdolomit", which also gives the northern border of the basin. The oldest members of the basement (Silurian and Carboniferous) can also be found in uplifted position in surface exposures (Szárhegy, Füle, Balatonfőkajár), and provide the souther boundary of the basin. The basin is filled by Tertiary sediments, of which the Eocene is subordinate, while the Miocene is nearly complete. From the five Miocene stages only the Badenian is discussed in detail, since only this stage has a closer relationship with oil shale formation. The discussion is based mainly on the references given by KÓKAY [1978]. In the Várpalota basin these Badenian sediments overlie the Carpathian Stage and can be subdivided into three stragitraphic units.

In the Lower Badenian (Moravian) marine sediments were deposited, in which coarse-grained sandstones are frequent in the western part of the basin, while clays and fine grained sandstones are predominant in the eastern part. It is probably interesting that very few volcanic tuffs and bentonites have been found in the Lower Badenian sequence of Várpalota compared to the formations in the Herend-Márkó basin, which belongs to the same stage. The average thickness of the Lower Badenian sediments, including the sands exposed in the famous Szabó-quarry, varies between 20—40 m.

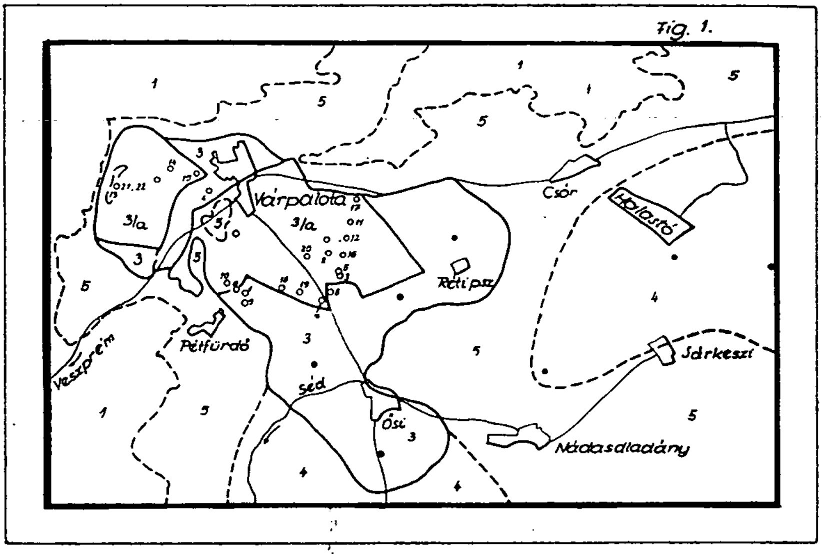


Fig. 1. Prognostic map of the oil-shale formation of the Várpalota basin
Scale 1:100 000
LEGEND
1. Surface exposures of the formations underlying the diatomaceous clay marls.
2. Surface outcrops of the diatomaceous clay marls.
3. Sub-surface extension of the diatomaceous clay marl.
3/a. Diatomaceous clay-marl in the hanging-wall of striped-out coal-seams.
4. Possible extension of diatomaceous clay marls overlain by younger formations.
5. Areas where diatomaceous clay marl does not occur.
014. Location of samples
● Proposed drill holes.

Following an uplift at the end of the Early Badenian the Mid- to the Late Badenian sedimentation was introduced by a transgression, or more accurately, expansion of the sea [KÓKAY, 1978]. This new cycle started with development of swamps and lignite formation. As a result of the extensive development of swamps, lignite was formed over an area of 60 sq. km. On the top of the shallow swamp facies pink rhyolite tuffs fell into seawater, producing the so-called "middle strip". These tuffs provided an evidence of volcanic activity associated with moderate structural movements. This pink rhyolite tuff bed grows thicker northwestwards and attains 2—3 m in thickness, indicating an increasing rate of land-derived, terrigene supplies.

The lignite seam is overlain by fossil-rich sediments containing *Neritina, Congeria, Teodoxus* and *Bulimus* in a thickness of few cm-s to several ten cm-s. This fossils often forms a lumachelle bed. With continued deepening of the lagoon the number of the Molluscs disappear. These layers represent the immediate foot-wall of the oil-shales of Várpalota.

In the Late Badenian the depth of the Várpalota Lagoon was at least 20 m. Laminated clay marl overlie the mollusc-rich hanging wall of the coal measure. The thickness of these strata is approximately 8—10 m. The rocks show greenish grey, green, occasionally brown colours and intense lamination characterized by alternation of very thin Ca-rich and clay laminae, which also contain some fossilized remnants of fish and produce a typical though slight, "oil-shale" odor when dried. This micro-bedding is probably due to seasonal changes [J. KÓKAY, 1966].

The lower diatomaceous clay marl series is overlain by rhyolite tuff throughout the basin in about one meter thickness. This tuff layer also shows a gradual thickening towards the peripheral parts. It is of loose sandstone character, being moderately sorted. Its bottom part is relatively coarse-grained and shows a gradual decrease in grain size upwards. Lapilli of 1 cm in diameter were occasionally found. In some parts it shows varying degrees of subsequent alteration to bentonite.

Absolute age determination of the rhyolite tuffs was made by using the K/Ar method [K. BALOGH *et al.*, 1979]. The most complete analyses were made on those samples which have been collected at Bántapuszta. The results obtained for different fractions did not exceed the limits of standard deviation. The average age, as determined by 3 measurements, is 14.6 ± 0.4 m. y.

Above the tuff layer the deposition of the diatomaceous clay marls continued. The colours of the rock change to monotonous grey, and its laminated character gradually disappears. It shows thicker parting, the typical oil-shale odor is much weaker, the diatomite micro-layers are not abundant, and the unit weight of the rocks becomes similar to that of typical clay-marls.

The laboratory analyses for organic matter and the Fischer-tar-content (performed in the MÁFI and the MÁFKI) supported the conclusions of field observations, which had indicated the highest grade of oil shales immediately below the tuff layer in few m thickness. According to their lithological character, these rocks are called „paper-shale". Like in the case of the oil-shales formed in crater-lakes, we have found evidence in Várpalota, too, that for the formation of oil-shales in confined landlocked basins and lagoons the existence of volcanic glass, which can easily devitrify, seems to be favourable.

Above the diatomite-clay marl series the sediments with molluscs *(Bulimus, Teodoxus)* reappear. This lithology indicates a regression, and accumulation. The disappearance of *Congeria* bears witness to a decreasing salinity.

The end-member of the sedimentary cycle is a coal-bed of high clay content and 1—2 m thickness [KÓKAY, 1978].

Significant differences from this typical sequence are known on the edges of the basin, the central part is filled with a monotonous diatomaceous clay-marl series, containing only one thin rhyolite tuff intercalation. Its maximum thickness, 142.9 m, was recorded in the DDH V-271, west from Várpalota. The average thickness in the basin is approximately 50 m.

The Badenian sediments are overlain by terrestrial pebbly variegated clay in the western part of the basin, in the eastern part Sarmatian oligohaline clay-marls and fine-grained clays are deposited, which are covered by a homogenous Lower Pannonian clay-marl series. On the top of the stratigraphic column there are Pleistocene and Holocene sediments.

Apart from a few local disturbances the foot-wall surface of the diatomaceous clay-marl series (oil-shale) slopes steadily eastwards. The thickness of the overlying strata varies between 0—313.6 m, and shows and eastward increase. The variation in the thickness of the diatomaceous clay-marl does not seem to be related to the paleogeographical environment of the foot-wall formations or the thickness of the overlying strata. This is mainly due to the unconformity between the Badenian and the Sarmatian and the erosion that followed by development of diatomaceous clay-marls and preceded the deposition of the Sarmatian sands and gravels.

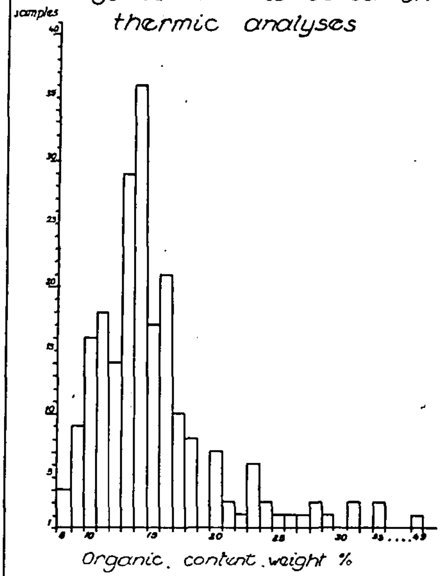
The best exposure of the diatomaceous shale (oil-shale) can be seen in a vertical section in the Bántapuszta open pit.

In addition to numerous drill hole intersections this formation is also known from underground workings, being the immediate hanging wall of the coal measure and usually hoisted to the surface and dumped as waste.

THE ORGANIC- AND CARBONATE CONTENT OF THE OIL-SHALES

Approximately 200 samples of oil-shales from 22 profiles in the Várpalota basin were investigated by thermal analysis. These measurements were performed by M. FÖLDVÁRY. The organic content of these samples was measured and found to vary between 8 and 49 weight %. The average organic matter content for the whole diatomaceous clay-marl series of the Várpalota basin is 15.33 weight %. A typical three-maxima distribution is seen on both organic content and carbonate content frequency diagrams of the diatomaceous clay-marl samples, according to the thermal results *(Fig. 2)*.

Fig. 2. Frequency distribution of the organic contents based on thermic analyses



The three maxima, however, represent only poor concentration of these values since both the organic and the carbonate contents vary widely and even the "striking" peaks with 35 values form only 17.5 per cent of the total number of analyses. This implies at the same time an ample scale of variations from 8 to 48 weight % values in the case of organic matter and from 0 to 90% in that of carbonate. The calcite and organic matter of the diatomaceous clay-marls of Várpalota, as determined by thermal analyses, do not show any correlation with each other.

The Fischer-tar-analyses have shown convincingly that the diatomaceous clay-marl sequence in the Várpalota basin is an oil-containing rock which includes single layers of excellent quality, but which, in sum total, can be rated as a low-grade oil shale *(Figs 3, 4)*.

Among all the samples analyzed, the lowest tar content, 0.3 weight %, was recorded in a sample from DDH V—82, from a depth of 156.6—171.0 m. Remarkably enough, 17 weight % organic matter belongs to this tar content. With similar amount

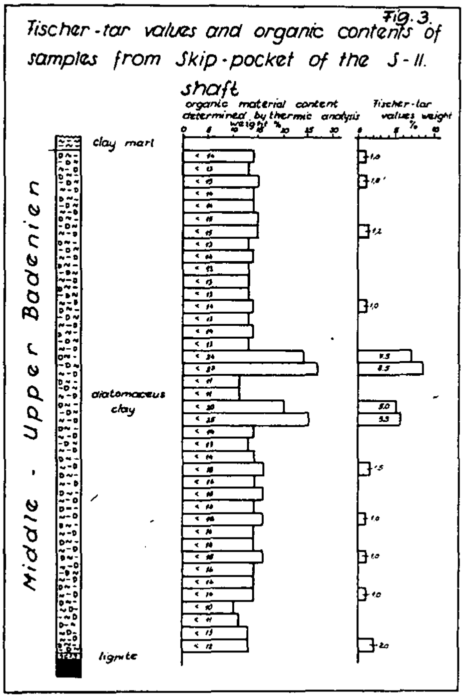


Fig. 3. Fischer-tar values and organic contents of samples from Skip-pocket of the S-II. shaft

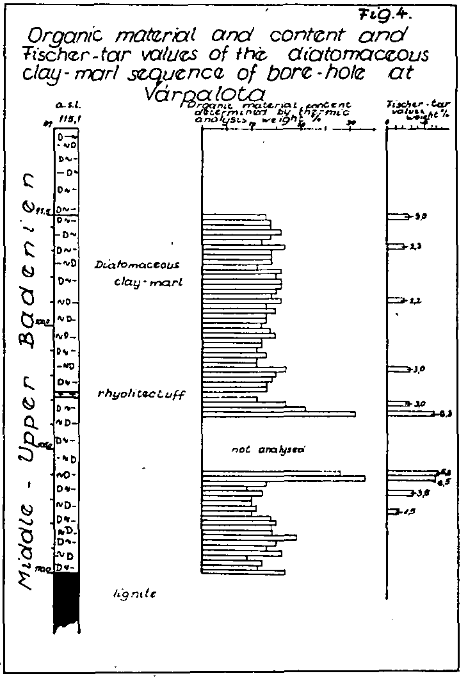


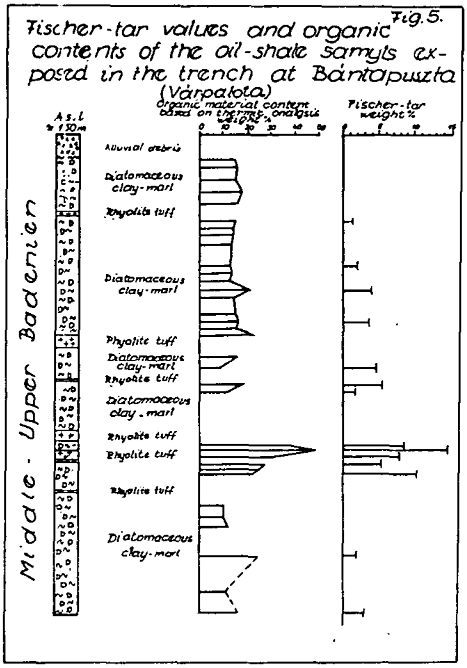
Fig. 4. Organic material and content and Fischer-tar values of the diatomaceous clay-marl sequence of bore-hole at Várpalota

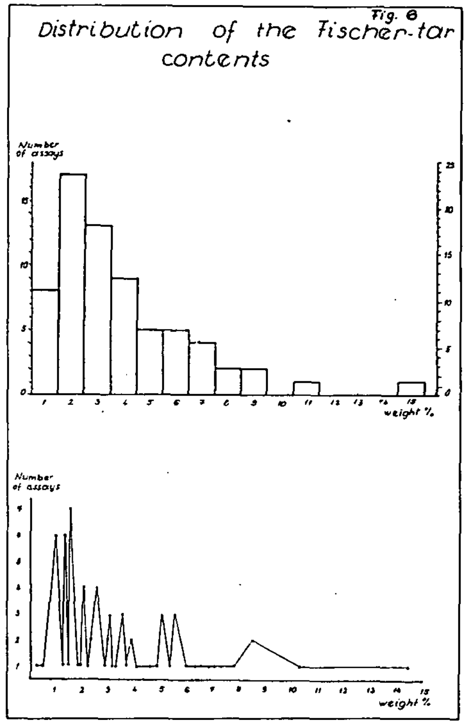
of organic material, a sample from a drillhole V—75 108.0—113.43 m yielded 5.0 weight % shale oil. The maximum tar content was found in a sample from a test trench at Bántapuszta *(Fig. 5)*. In this sample 49 weight % organic matter (which als represented a maximum) and 14.5 weight % tar were measured (160 l/ton shale oil). For those 71 samples, which contained at least 9 weight %, but mainly 15 weight % organic matter, the average Fischer-tar-content was 3.4 weight % (37 l/ton). These values fall in the range of low grade oil shales. As is shown in the distribution diagram of the Fischer-tar-content of the samples *(Fig. 6)* the modal mean value has fallen in the range of 1—2 weight % tar concent. About one fourth of the total number of samples (17) fit into this interval.

36 samples had a tar content of 2—7 weight %, over fifty per cent of the total number of samples.

The already known close correlation between the organic material content of the oil-shales determined by thermal analysis and their Fischer-tar-content is well see in the correlation diagram of these samples too.

The 71 analyses are sufficient for a statistical evaluation. It should be mentioned that these data are only informative, since only those samples have been analyzed for oil content, which had shown at least 8 weight % or rather over 15 weight % organic matter. The correlation index for the organic content and Fischer oil-yield is r=0.848, marking a good correlation. The ratio of organic matter to tar is 18.239: :3.386=5.4:1, which means, that about 18.5 per cent of the organic matter can be converted into tar (shale-oil) by Fischer's distillation techniques. If we compare this





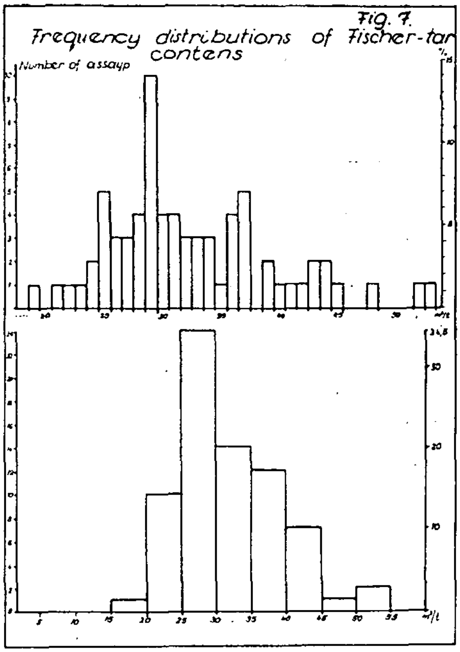
value to the 15.3 weight % average organic matter content of the analyzed 200 samples, we get 2.74% as an average Fischer-tar-content. The relationship between organic matter and tar in the 71 samples is expressed by

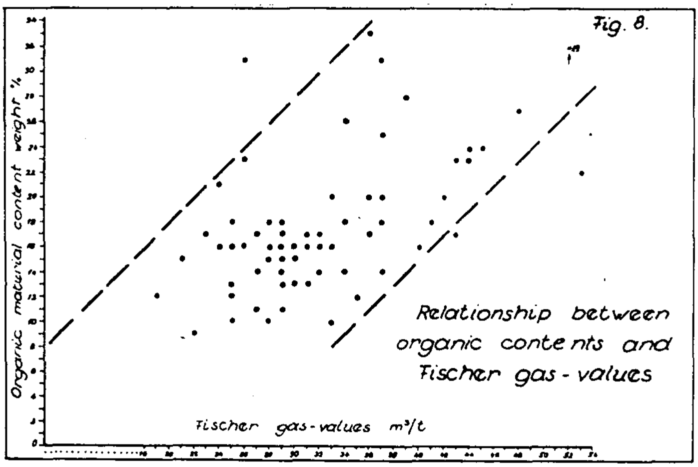
$$x=\frac{x}{3}-1$$

while the total organic matter and the tar contents show a correlation, which can be described by

$$y=0.1944\,x$$

Apart from the recoverable oil content, the most important property of these oil-shales is their gas yield of low calorific value during Fischer distillation. The 71 samples produced 32 m³/ton average amount of gas. Although this gas content is not in a genetic relationship with the tar content, the results have shown that those samples, which have a higher tar content, yield a larger amount of gas. The maximum amount of gas (53 m³/ton) was recorded at 3.6 weight % tar content, but those samples having 14.5 weight % tar also produced 52 m³/ton gas. At the minimum — 19 m³/ton — gas yield, the tar content was 1.3 weight % in the sample (*Figs. 7, 8*).





The analyses for coal quality of 4 samples from the Várpalota basin have given suprising results. The hanging-wall rock, which was hitherto considered a waste, has a calorific maximum vaule as 10 275 kJ/kg with 10.3% tar, 59% ash and 4.5% moisture. (Table 1.)

TABLE 1

Section No	Locality	Quality control (MEO)				Fischer-tar weight%				By derivatographic method					
		Calorific power kcal/kg	Calorific power kJ/kg	Ash content A %	Moisture wt %	Volatile content	Tar	Underground moisture content	Moisture	Water as a decomposition product	Waste + koke	Gas and Losses	gas m^3/t	Organic matter content	$CaCO_3$ content
13	Exploratory trench. lager 20	2454,1	10275	59,41	4,53	29,84	10,3	7,9	6,6	2,2	77,5	3,2	43	23	20-25
15	Face 45 in Bántabánya mine	798,7	3344	38,16	21,76	16,70	5,0	21,9	6,6	0,9	86,5	1,0	44	23	72-73
16	Sampling for Skyp S-II	1159,1	4853	67,50	5,51	24,62	8,5	11,8	5,4	1,6	82,0	2,5	48	27	9-10
17	S-II mine 12/A face	369,7	1548	42,29	19,41	16,81	3,2	21,1	3,9	0,4	90,6	1,9	40	16	57-58

Palynological analyses were made in 27 samples from the Várpalota basin by MRS. L. NAGY and E. BODOR. The results of these analyses have supported the relationship of these strata with the Hidas Formation, with *Sporomorphae* indicating Middle Miocene age.

The pollen content has not allowed any more accurate subdivision within the Badenian stage. In a sample from V—82 drillhole, 205.0—214.85 m section, *Cyrillaceae, Savalpollenites* and *Ophioglaglossissporites* pollen grains, indicating the Lower Badenian, were observed.

In the DDH V—75, 65.2—98.7 m section the samples contained fewer tropical to subtropical elements, and an increasing number of arcto-Tertiary elements, an assemblage, which marks the upper horizons of the Badenian. Unlike the other freshwater *Sporomorphae* assemblages, these samples are of marine character, along with a large number of *Hidasia sp.* plankton specimens. This planktonic genus did not occur in other samples. Because of the relatively high abundance of *Coniferae*-pollenites in the samples, the climate of this area seems to have been moderately warm, subtropical during the Middle Badenian.

The character of vegetation shows an environment near to freshwater sources *(Myrica, Carya, Pterocarya)*, swamp forests *(Taxodium)*, and mainly wet deciduous forests *(Ulmaceae)* with a dense undergrowth near rivers and stream banks. The *Coniferae, Fagaceae* and *Ulmaceae* assemblage belongs to the vegetation of cooler mountain slopes. Local predominance of pine forests was indicated by a few samples (DDH V—128, 207.0—210.3 m).

TABLE 2

DDH	m	Amount of *Botrycoccus*	Org. mat. content (weight %)	Fischer tar content (weight %)
V-75	65.2— 98.7	rare 1—3	<16	1.3
V-82	205.0—214.85	none	<11	
V-90	173.0—209.9	none	<22	3.6
V-128	181.1—206.0	much 11—15	<17	1.4
V-128	207.0—210.3	dominant 16—00	<11	
V-122	73.2— 98.8	rare 1—3	<18	3.1
V-212	167.8—190.5	dominant 16—00	<15	2.3
V-212	206.0—207.0	none	trace	
V-317	95.5— 95.7	rare 1—3	<13	3.0
V-317	96.1— 96.3	few 4—5	<16	
V-317	96.7— 96.9	rare 1—3	<17	2.3
V-317	97.3— 97.5	none	<14	
V-317	97.9— 98.1	much 11—15	<15	
V-317	98.9— 99.1	much 11—15	<16	2.2
V-317	99.1—100.1	rare 1—3	<12	
V-317	101.9—102.1	few 4—5	<15	
V-317	102.9—103.1	much 11—15	<11	
V-317	103.5—103.7	rare 1—3	<31	6.3
V-317	106.9—107.1	dominant 16—00	<10	
V-317	107.9—108.1	rare 1—3	10—15	
V-317	108.9—109.1	dominant 16—00	9—13	
V-317	109.9—110.0	rare 1—3	<17	
I-13	157.0—160.0	none	<18	
S-II	shaft, reject-dump	dominant 16—00	<16	
S-II	shaft	much 11—15	<14	

THE ALGAL CONTENT OF THE OIL SHALE SAMPLES

Whether an organic-rich rock qualifies for oil shale or not, depends to a large extent on the amount of the accumulated *Botryococcus* algae, which are actually hydrocarbon producers.

In the 27 analyzed samples from Várpalota, only five contained significant amounts of *Botryococcus braunii* algae, which account for the kerogene content of the oil-shales. There were 6 samples, in which no algae were found. In the remaining 16 preparates the number of the algae varied from 15 to 1.

The relationship of the abundance of *Botryococcus versus* organic matter and the Fischer-tar-content of the samples is shown in Table 2.

Suprisingly enough, the data shown by Table 1, indicate a negative correlation between the amount of *Botryococcus* and the organic matter content of the rocks determined by thermal analyses. At 17 weight% organic matter there were only 1—3 specimens of *Botryococcus braunii*. In those samples, in which *Botryococcus* was abundant, the amount of organic matter was less than 16 weight %.

M. Hajós has studied the *Diatomae* in the samples from drillholes in Várpalota. She found that the *Diatomae* content was very diffeıent from drillhole to drillhole, and even within the section of one drillhole. Limno-brackish and euryhaline genera are predominant in the assemblage. Typical representatives of the normal marine environment are absent.

CONDITIONS OF DEPOSITION

The diatomaceous sediments of the basin are very rich in carbonized plant fossils, though contain only small amounts of *Diatomae* and other siliceous fossils. The site of sedimentation was a basin, relatively deep, with plankton-rich waters, in a regime of gradual accumulation, upfilling. The diatcm assemblage is relatively simple, with only few genera, mainly planktonic types. This marks a lake-type sedimentation, forming a relatively smooth, muddy bottom.

The variability of the abundant halophytic *Coscinodiscus species* shows not only a decrease in.salinity, but its constant fluctuations as well. Most of the other genera are also typical of limno-brackish, or freshwater environments. However, the typical freshwater elements aıe subordinate.

The molluscs, the rich pollen and spore remains, and mainly the planktonic diatoms undoubtedly indicate low salinity.

Using Remane's classification, the salinity falls to the meso-haline range, with up to 5—7% concentration.

Finally, the *Diatomae* have again proved a Middle Miocene Badenian age of deposition.

ECONOMIC SIGNIFICANCE OF THE OIL SHALES

For an estimation of the approximate hydrocarbon reserves contained in the diatomaceous shale series of the Várpalota basin the following data can be considered.

The extension of the formation is about 50 sq. km; its average thickness is 50 m; its unit weight is approximately 1 g/cm^3. Calculating with these values we have obtained the results:

Possible shale oil reserves (2.7 weight %): 68 million metric tons;

Possible shale gas reserves (32 m^3/ton): 80 billion m^3.

Further subdivision of these reserves has not been possible in this exploration stage.

The 68 million tons of shale oil reserve are stored in low grade oil-shales. With present-day technologies and market situation its exploration is economically unfeasible. It should be mentioned, however, that in 1965 the lowest limit of the economic grade of oil shale was about 10 weight %. Recently in-situ production from oil-shales is known from as low as 8 weight % grade oil-shales. There are also reasonable possibilities for the use of this raw material as an additive to manufacturing cement and fibrous insulating materials, according to the specialists of the Hungarian Reserarch Institute for Mineral Oil and Gas (A. FEHÉRVÁRI, J. BARLAI and G. KOCSIS).

The oil shale series of Várpalota has a favourable setting for the mine development. In the Bántapuszta area it has been already exposed in a face of several hundred m length with 1—4 m overburden. Here conventional strip mining of several ten million m^3 of oil-shale is possible, despite the increasing thickness of the overburden eastwards, since the analyses have shown gradually better grades in that direction.

Other deposits are not accessible to surface mining methods owing to higher overburden thicknesses.

Since underground coal mining is in operation in the area, underground technologies may also be considered for the extraction of the oil-shales using the same methods and equipment and labour as for coal. Along with the coal production up to 1—2 millions tons of oil-shale can also be hoisted. Since a large amount of this material is in any case hauled from the mine as waste, its processing as oil shale should be seriously considered. In judging the future perspectives of coal mining in Várpalota, the diatomaceous shale overlayers, which may yield hydrocarbon, should also be taken into account in the development plans, as the oil prices show a trend of steady increase in the world market.

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