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HYDROCARBON GENERATIVE FEATURES OF THE UPPER TRIASSIC KÖSSEN MARL FROM W. HUNGARY

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ABSTRACT

The organic matter of the Kössen Marl accumulated in an anoxic environment is supposed to be an oil prone sequence. A verification of this assumption is attempted in this paper by determining its organic facies and its petroleum potential.

The relationship lithology and hydrocarbon generative features of the Kössen Marl was studied. One part of the samples was chosen from a shallow corehole consisting of mainly dolomite. The other part of the samples originated from three subdivisions of a corehole drilled in the vicinity of the previous one. The subdivisions were the following: calcareous marl, marl and silstone.

The petroleum potential and the hydrogen index of the dolomite indicated a good source rock of Organic Facies B. The calcareous marl proved to be a typical example of the excellent oil producing matter. It had high petroleum potential and its organic facies was nearly AB. The marl also seemed to be a very good source rock. However, its petroleum potential was less than half of the value measured in the previous subdivision and its organic facies was BC. The siltstone of Organic Facies D was found to be a nongenerative rock.

The hydrocarbon generative features mentioned above were checked on two selected samples by simulating the catagenesis under laboratory conditions by performing a thermal degradation.

KEYWORDS: hydrocarbon generative features, kerogen types, organic facies, petroleum potential, source rock, thermal degradation

INTRODUCTION

The Upper Triassic Kössen Marl from West Hungary is considered to be a good oil source rock. A relationship is supposed to exist between its organic matter and the Nagylengyel oil field which contains heavy and sulfur-rich crude oil. On the basis of sulfur content, stable isotope ratios and trace elements KONCZ (1984) suggested that there is a Triassic carbonate source rock for the crude oil of the Nagylengyel field. Two shallow coreholes located in the vicinity of this oil field — Rezi 1 (referred to as R) and Zalaszentlászló 1 (referred to as Z) — were studied by BRUKNER-WEIN and VETŐ (1985). They investigated the relationship between lithology and organic matter in anoxic sediments. Results of their work — organic carbon content, the quantity of iron and that of different forms of sulfur, as well as detailed geochemical analysis of extracts — indicated that the examined sequence of Kössen Marl is an excellent oil source rock. The fine-laminated structure and the absence of benthic remains were evidences of an anoxic depositional environment.

Microscopic examination of the amorphous kerogen revealed that the precursor biomass was algae and associated bacteria in various stages of decomposition In core R Botryococcus remnants could be identified. In core Z strongly degraded palynomorphs were found (GÓCZÁN, unpublished data).

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The conclusions of works mentioned above could be confirmed further by some data concerning the type of kerogen, the organic facies and petroleum potential. The aim of the present paper was to determine these geochemical parameters of the selected sequence of Kössen Marl, as well as to study in detail its hydrocarbon generative features by thermal degradation.

Simplified lithologic logs of the Kössen Marl in wells R and Z (after BRUKNER-WEIN and VETŐ, 1985) are shown in *Fig. 2.* Lithology of the sequence selected for study varies from carbonates to siltstones. The carbonate mineral in core R is mainly calcite, in core Z is mainly dolomite.

EXPERIMENTAL

The total organic carbon content (TOC) was measured at 1000 °C under intense oxygen flow by combusting in Carmograph—8 equipment.

The hydrocarbon generative (petroleum) potential, the maturity of the kerogen the hydrogen- and oxygen-indices were determined by Rock Eval pyrolysis (ESPITALIÉ *et al.*, 1977).

Experimental assay of thermal evolution was carried out in a temperature-programmed furnace under nitrogen atmosphere. Temperatures of the experimental evolution were 350°, 375°, 400°, 450° and 500° C. At each temperature the heating period lasted for 1, 5, an 10 hours. Volatile bitumens were collected in cooled traps, soluble bitumens were extracted by chloroform. The residue of thermal degradation (unconverted kerogen) was characterized by Rock Eval pyrolysis and by CR/CT ratio measured according to the ASTM standard (CUMMINS *et al.*, 1972).

RESULTS

1. *The quality of organic matter*

The quality of organic matter was characterized by the organic facies and the type of kerogens. “The organic facies is specific body of sedimentary rocks identified by the same organic features” (JONES, 1987). The same organic facies may contain a variety of kerogen types in different mixtures. One of the bases of classification is the hydrogen index measured at a vitrinite reflectance $\simeq 0.5\%$. Immaturity of the kerogen is an essential condition for determining the organic facies. As it is shown in Table 1 and 2, as well as in *Fig. 1* maturity of the organic matter in each examined sample meets this requirement. So the maturity level of kerogens in both of the coreholes is the most favourable to state their type and organic facies, as well as to compare them to each other.

The type of kerogen. Nearly all the types of kerogen can be found among the 72 examined samples *(Fig. 1).* The hydrogen indices change from 38 mgHC/gTOC (type III) to 968 mgHC/gTOC (type I). On the basis of the average value of the hydrogen indices both wells contained kerogen of type II. In corehole Z the average HI was 413 mgHC/gTOC while it was 454 mgHC/gTOC in corehole R. The PC/TOC ratios were also very characteristic of the kerogen of type II (36% and 40%, respectively).

In corehole Z almost each core contained kerogen of type II (Table 2 and *Fig. 1*).

In contrast, the quality of organic matter in three subdivisions of corehole R was very different *(Fig. 1* and Table 1*)*. In calcareous marl, although the kerogen

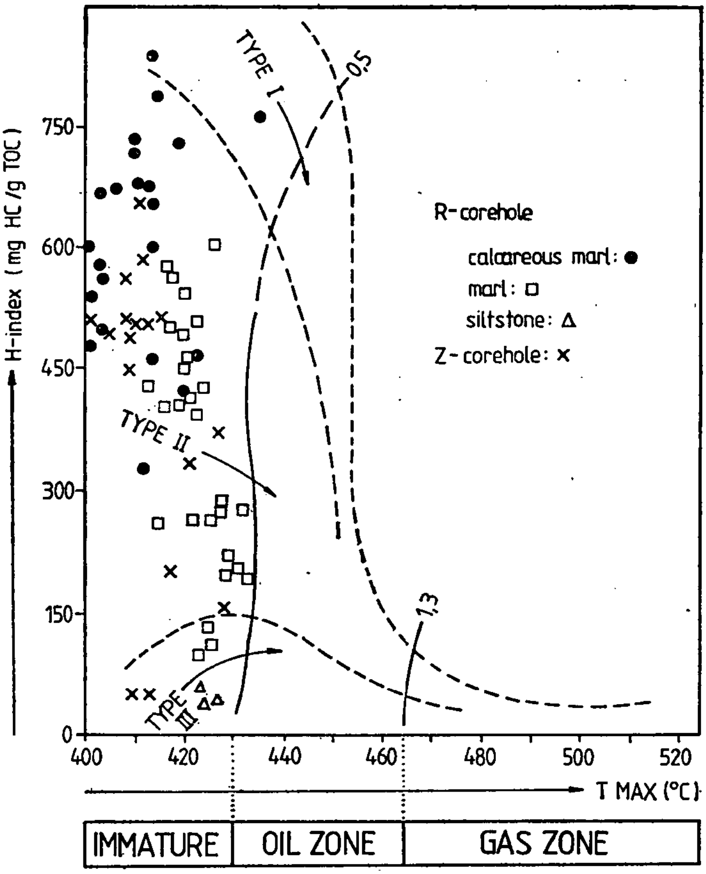

Fig. 1. HI – T_{max} diagram showing type and maturity of kerogen of Kössen Marl in wells R and Z

was type II, the values of HI and PC/TOC ratio approached the boundary between the kerogen of type I and II. Most of the studied kerogen in this section proved to be pronouncedly type I—II (Table 1).

The kerogen of marls changed from one type to another at a depth of about 110—120 m (dotted line in Table 1). The average value of HI decreased from 444 mgHC/gTOC to 198 mgHC/gTOC and the PC/TOC ratio decreased from 39% to 17%.

Not only the quantity, but the quality of the organic matter of siltstone turned out to be very poor. These kerogens showed low HI and low PC/TOC ratio, too.

Organic facies. The general organic facies of both examined wells is B. The organic facies B is considered to be the predominant source of the world's oil. The precursor biomass of the examined Kössen Marl consisted mainly of Botryococcus algae and associated bacteria in various stages of decomposition (GÓCZÁN, unpublished data). However, some terrestrially derived organic matter varying in amount and in type was present, too. Depending on the ratio of aquatic and terrestrially

TABLE 1

Organic geochemical characterization of core Rezi 1 (southwest part of Transdanubian Mid-Mountains, Hungary) by Rock Eval pyrolysis

Depth (m)	TOC (%)	T_{max} (°C)	HC-pot kgHC/ton of rock	S2/S3	PC/TOC (%)	HI mgHC/g TOC	OI mgCO_2/g TOC	Type of kerogen	Org. fac.
1	2	3	4	5	6	7	8	9	10
44.0	0.84	426	0.37	0.85	3.57	42	50	III	D
49.1	0.78	424	0.30	0.90	2.56	37	41	III	D
59.8	0.72	423	0.39	1.00	4.17	52	52	III	CD
82.4	0.67	432	1.94	2.30	23.88	279	120	II	BC
88.4	1.06	425	1.56	2.22	12.26	138	62	III	C
88.6	1.51	433	2.95	4.11	15.89	190	46	II	w
92.6	1.06	426	1.16	1.92	8.49	103	53	III	CD
93.6	1.17	423	1.20	1.73	8.55	93	53	III	CD
99.8	1.72	431	3.58	5.17	16.86	201	38	II	C
101.3	1.92	429	4.30	4.90	18.23	217	44	II	C
102.6	1.39	428	2.79	2.65	16.55	194	73	II—III	C
107.6	1.83	427	5.21	3.95	23.50	276	69	II	BC
113.2	1.62	427	4.80	3.92	24.70	288	73	II	BC
122.6	4.76	426	29.96	16.52	51.89	607	36	II	B
129.6	3.46	420	19.27	9.54	46.24	540	56	II	B
136.6	5.01	417	26.70	13.06	44.31	503	38	II	B
147.6	3.13	420	16.41	9.54	43.45	494	51	II	B
148.6	7.92	416	48.65	19.67	51.14	578	29	II	B
149.6	1.94	424	8.79	5.54	37.63	422	76	II	B
151.6	6.08	423	33.54	17.57	45.89	517	29	II	B
156.6	3.72	419	15.63	9.77	34.95	401	41	II	B
162.5	5.53	417	32.13	15.61	48.28	558	35	II	B
171.6	3.03	421	14.97	9.80	40.93	468	47	II	B
175.5	1.66	426	4.45	3.82	22.29	257	67	II	BC
177.5	2.25	415	6.42	4.15	23.56	271	65	II	BC
179.5	3.55	423	14.94	9.06	34.93	403	44	II	B
180.5	3.41	421	15.19	8.14	36.95	422	51	II	B
181.5	2.66	418	11.10	7.83	34.59	400	51	II	B
187.5	2.76	420	13.14	9.04	39.50	458	50	II	B
190.5	1.62	422	4.34	3.50	22.22	254	72	II	BC
192.5	2.15	413	9.90	5.83	38.14	433	74	II	B
202.7	2.69	419	21.53	11.53	66.54	737	63	I—II	AB
211.9	3.73	404	22.74	10.58	50.67	570	53	II	B
213.8	10.34	415	86.84	25.97	69.92	788	30	I—II	AB
215.9	4.43	414	39.59	11.99	74.27	833	69	I	AB
216.6	8.65	414	56.73	17.09	54.56	604	35	II	B
217.4	6.05	436	62.82	18.65	86.44	968	51	I	A
218.0	4.02	414	28.43	9.37	58.70	653	69	I—II	AB
220.9	4.12	423	19.88	8.26	40.04	469	56	II	B
223.8	2.72	420	11.76	7.58	36.02	420	55	II	B
225.8	8.57	412	31.54	8.02	30.57	326	40	II	BC
227.4	5.59	410	45.71	17.30	67.98	740	42	I—II	AB
229.5	4.01	407	32.04	17.42	66.58	715	40	I—II	AB
230.5	5.76	413	42.04	15.19	60.70	672	44	I—II	AB
234.7	8.60	411	33.94	11.69	61.30	676	57	I—II	AB
235.7	2.72	401	16.08	8.52	49.26	535	62	II	B
240.4	2.90	401	15.45	5.22	44.14	479	91	II	B
241.4	6.49	400	42.46	15.72	54.39	608	38	II	B
242.0	6.58	403	40.20	14.39	50.91	568	39	II	B
250.0	2.97	414	14.77	6.56	41.41	464	70	II	B
251.6	2.04	403	15.56	8.42	63.23	664	78	I—II	AB
253.6	3.60	407	27.41	11.02	63.33	670	60	I—II	AB
266.3	2.60	436	20.21	10.48	64.61	762	72	I—II	AB
269.0	2.81	404	15.28	8.71	45.19	499	57	II	B
Average values	3.57	411	20.54	9.13	40.31	454	55	II	B

derived organic matter, different organic facies could be recognised in the two wells and in the three subdivision.

In the corehole Z there were two samples of Organic Facies D. Their organic matter consisted primarily of inertinite maceral groups and this facies is non-generative. Except for these two cores the organic facies of almost each sample was B (Table 2).

In corehole R the organic facies changed with the depth. The oil prone Organic

TABLE 2

Organic geochemical characterization of core Zalaszentlászló 1 (southwest part of Transdanubian Mid-Mountains, Hungary) by Rock Eval pyrolysis

Depth (m)	TOC (%)	T_{max} (°C)	HC-pot kgHC/ton of rock	S2/S3	PC/TOC (%)	HI mgHC/g TOC	OI mgCO$_2$/g TOC	Type of kerogen	Org. fac.
1	2	3	4	5	6	7	8	9	10
304.9—307.0	31.5	412	22.53	16.09	59	653	40	I—II	AB
317.6—318.4	4.56	408	25.09	19.40	46	510	26	II	B
322.0—325.0	0.17	427	0.67	0.85	29	370	435	II	BC
337.6—340.7	1.11	416	5.90	5.56	44	511	91	II	B
341.5—344.0	0.17	417	0.36	0.70	18	200	282	II	C
349.1—352.0	2.25	405	11.74	13.85	43	498	36	II	B
354.8—360.0	2.48	412	15.36	19.79	52	590	29	II	B
360.2—363.0	2.25	413	11.78	15.01	44	507	33	II	B
363.0—366.8	5.23	408	30.33	23.38	48	554	23	II	B
375.0—378.5	2.09	401	11.23	15.22	45	510	33	II	B
378.5—381.4	2.98	410	15.62	22.75	44	504	22	II	B
381.4—383.0	2.45	409	12.65	20.33	43	489	24	II	B
383.0—386.0	0.72	409	3.37	6.69	39	455	68	II	B
387.0—388.0	1.85	413	9.88	14.33	44	507	35	II	B
407.8—408.8	0.86	421	3.02	3.05	29	337	110	II	BC
410.9—413.0	0.65	428	1.05	1.36	12	155	113	II—III	C
421.5—425.5	0.33	413	0.14	0.30	3	42	139	III	D
434.0—436.8	0.40	409	0.24	0.25	5	47	187	III	D
Average values:	1.87	413	10.05	11.06	36	413	166	II	B

Facies AB and B were followed by mixed Organic Facies BC, then by the gas prone Organic Facies C and finally, by the non-generative Organic Facies D (Table 1).

The average HI of the calcareous marl reached almost the beginning of the Organic Facies AB (HI=650 mgHC/gTOC). Numerous samples even exceeded this value. Within the marls of Organic Facies BC two sections could be distinguished Organic Facies B (HI=444 mgHC/gTOC) and Organic Facies C (HI=198 mgHC/ /gTOC).

2. *Source rock considerations*

The organic carbon content and the petroleum potential of the examined samples indicated that the Kössen Marl was a good source rock. Their organic carbon content ranged from 0.2% to 10.3 and their petroleum potential varied from 0.3 kgHC/ton of rock to 86.8 kgHC/ton of rock *(Fig. 2)*.

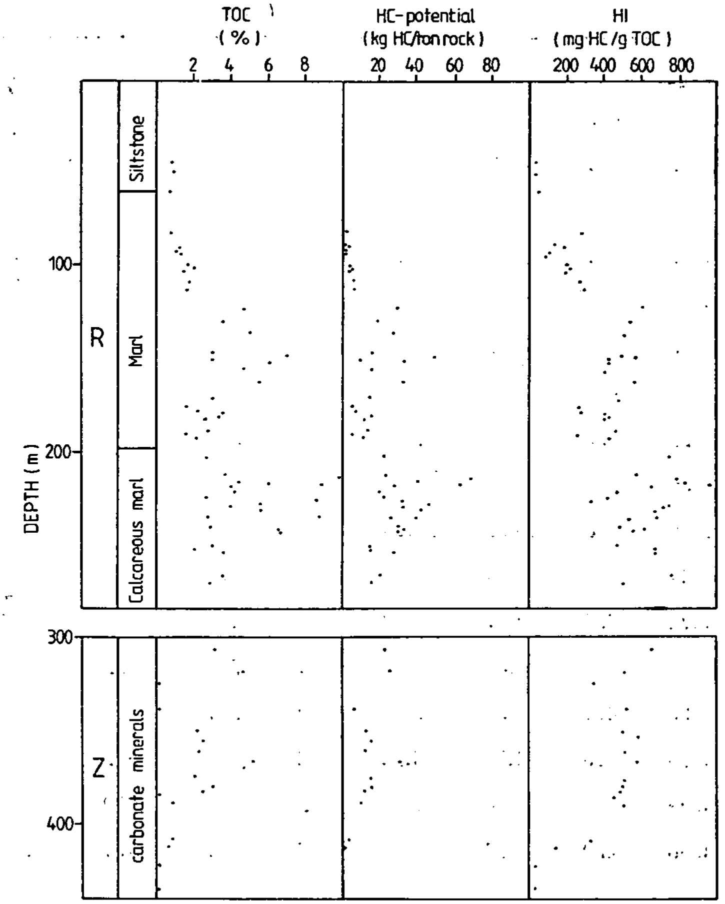

Fig. 2. Simplified lithologic logs of Kössen Marl in wells R and Z as well as hydrocarbon generative features vs. depth

On the average values of these parameters (Table 2) core Z of Organic Facies B proved to be a "good source rock". According to Peters (1986) the organic carbon content of a "good source rock" is 1—2% and its hydrocarbon generative potential is 6—12 kgHC/ton of rock.

The other studied core (R), which also contained Organic Facies B, seemed to be a "very good source rock" its TOC being higher than 2% and its petroleum potential higher than 12 kgHC/ton of rock (Table 1).

In addition to their average organic carbon content and their petroleum potential the high S2/S3 ratio of the calcareous marl (Table 3) also showed an excellent oil producing source rock.

On the basis of average values mentioned above the total section of marls also seemed to be a very good source rock (Table 3). However, similarly to the other geochemical parameters studied in this paper, the S2/S3 ratio also revealed two zones within the marl sequence. The average value of S2/S3 ratio (9.89) of the deeper zone of marl characterized an excellent oil producing source rock. On the other hand, this parameter (3.29) marked only a gas prone source rock in the upper zone (Table 1).

Siltstones containing a small quantity of organic carbon and being a non-generative facies cannot be regarded as a source rock.

Table 3

Average values of some quantitative and qualitative features of the organic matter in core Rezi 1

Lithological subdivisions	Depth (m)	TOC (%)	HC-pot kgHC/ ton of rock	S2/S3	PC/TOC (%)	HI mgHC/g TOC	Type of kerogen	Organic facies
Siltstone	44.0—59.8	0.78	0.35	0.92	3.43	44	III	D
Marl	82.4—192.5	2.81	13.04	7.53	30.92	356	II	BC
Calcareous marl	202.7—269.0	4.87	32.30	12.16	56.55	627	II	B

3. *Results of thermal degradation*

Experimental assay of thermal degradation was performed on two samples.

The core of Organic Facies B selected from well Z (depth=317,6—318.4 m) contained a very good oil producing kerogen. TOC=4.56%, HC-potential=25.09 kgHC/ton of rock, S2/S3=19.40, HI=510 mgHC/gTOC.

The core of Organic Facies AB selected from well R (depth=213.8 m) contained an excellent oil producing kerogen, too. TOC=10.34%, HC-potential= 86.84 kgHC/ton of rock, S2/S3=25.97, HI=788 mgHC/gTOC.

The quantity of total bitumens yielded by the two samples during the thermal degradation changed both in function of heating period and in function of temperature (Table 4).

The thermal degradation properties of core Z proved to be similar to those of the kerogen of type II. Evolution properties of the core R of high generating capacity, on the other hand, were revealed to be close to the kerogen of type I. A higher percent of the organic matter in sample R was converted to oil than in sample Z. Independently of the experimental conditions much more bitumen developed from sample R than sample Z (Table 4).

TABLE 4

Quantity of bitumens yielded by thermal degradation of two Kössen Marl samples (R: a sample of Organic Facies AB from core Rezi 1 and Z: sample of organic Facies B, from core Zalaszentlászló 1)

Temperature (°C)	Heating period (hours)	Soluble bitumen		Volatilized bit. mg/gTOC		Total bitumen	
		R	Z	R	Z	R	Z
350	1	222.4	197.4	106.4	43.8	328.8	241.2
	5	261.1	65.8	145.1	43.8	406.2	109.6
	10	299.8	43.8	193.4	43.8	493.2	87.6
375	1	164.4	65.8	116.1	43.8	280.5	109.6
	5	222.4	43.8	203.1	109.6	425.5	153.4
	10	251.5	21.9	270.8	109.6	522.3	131.5
400	1	348.2	197.4	145.1	109.6	493.3	307.0
	5	174.1	43.8	222.4	153.5	396.5	197.3
	10	280.5	21.9	560.9	175.4	841.4	197.3
450	1	154.7	43.8	435.2	131.6	589.9	175.4
	5	19.3	21.9	560.9	219.3	579.9	241.2
	10	48.4	21.9	647.9	175.4	696,3	197.3
500	1	19.3	21.9	647.9	263.1	667.2	285.0
	5	9.7	21.9	686.6	219.3	696.3	241.2
	10	9.7	21.9	715.7	285.1	725.4	307.0

TABLE 5

Change of some characteristics of unconverted kerogen type I—II (from core R) during the thermal degradation

Temperature (°C)	Heating period (hours)	CR/CT	T_{max}	S2/S3	HC-pot kgHC/ton of rock	HI mgHC/gTOC
Unheated sample		—	415	25.97	86.84	788
350	1	0.61	418	25.22	65.29	696
	5	0.66	422	24.05	51.43	695
	10	0.78	423	21.97	46.72	639
375	1	0.59	416	24.90	54.12	710
	5	0.74	424	21.23	38.82	573
	10	0.76	430	14.22	26.74	443
400	1	0.80	425	16.34	31.91	641
	5	0.87	436	7.51	16.45	382
	10	0.91	441	6.47	11.21	260
450	1	0.95	443	4.74	7.45	263
	5	0.96	455	1.17	2.21	70
	10	0.99	456	0.54	1.08	27
500	1	0.95	463	0.70	1.32	34
	5	0.98	>465	0.15	0.41	21
	10	0.99	>465	0.13	0.36	8

The kerogen of type I, however, requires higher temperature for generating oil than the other types. Moreover, the main formation of petroleum starts at a somewhat higher temperature (Tissot and Welte, 1978). As it is shown in Table 7 the hydrocarbon generating potential of sample R reduced to 1 % of its original value at 450 °C.

Table 6

Change of some characteristics of unconverted kerogen type II (from core Z) during the thermal degradation

Temperature (°C)	Heating period (hours)	CR/CT	T_{max} (°C)	S2/S3	HC-pot kgHC/ton of rock	HI mgHC/gTOC
Unheated sample		—	408	19.40	25.09	510
350	1	0.62	416	21.45	17.63	349
	5	0.88	426	8.75	8.13	172
	10	0.96	434	3.52	3.30	72
375	1	0.77	417	22.10	15.92	346
	5	0.85	436	6.27	4.54	101
	10	0.99	437	1.56	1.95	33
400	1	0.88	426	12.13	9.00	213
	5	0.93	440	1,78	2.64	42
	10	0.94	448	0.16	0.07	1
450	1	0.84	443	1.90	1.26	33
	5	0.93	>465	0,00	0.09	0
	10	0.93	>465	0.00	0.07	0
500	1	0.90	>465	0.03	0.15	0
	5	0.93	>465	0.10	0.18	0
	10	0.94	>465	0.00	0.09	0

Table 7

Decreasing of the hydrocarbon generative potential during the thermal degradation
(R: a sample of Organic Facies AB from core Rezi 1
Z: a sample of Organic Facies B, from core Zalaszentlászló 1)

Temperature (°C)	Heating period (hours)	Decreasing of HC-pot. %	
		R	Z
350	1	25	32
	5	41	68
	10	46	84
375	1	38	38
	5	55	82
	10	69	92
400	1	63	65
	5	81	90
	10	87	99
450	1	91	95
	5	97	100
	10	99	100
500	1	92	99
	5	100	100
	10	100	100

In contrast, residual petroleum potential of the sample Z, which required lower activation energy, was 1% at 400 °C. A significant difference was found between the two examined rocks regarding the decrease of their petroleum potential at the beginning of the catagenesis, which was simulated by thermal degradation at 350 °C for 10 hours. While the hydrocarbon generative potential of sample R fell to 56% of that of the unheated matter, the residual potential of sample Z was only 16%.

The so called "oil death line" (vitrinite reflectance = 1.35% and $T_{max} \geqq 465$ °C) was reached at 500 °C and at 450 °C by sample R and sample Z, respectively (Table 5 and 6).

CONCLUSION

1. On the basis of the results presented in this paper the Upper Triassic Kössen Marl (W. Hungary) had very good hydrocarbon generative features.
2. Both of the studied wells (R and Z) represented organic rich (TOC = 0.2—10.3%) shallow marine sequences. All types of kerogen occurred among the examined samples. In *Fig. 1* kerogen of type II in siltstone (core R) and in core Z could be recognized. The calcareous marl (core R) contained a kerogen of high quality, about type I—II.
3. The total zone of the catagenesis could be simulated by thermal degradation. This fact and the low T_{max} suggested that the organic matter was in the early stage of maturation
4. The ratio of the organic carbon content in the three lithological subdivisions of core R (siltstone, marl and calcareous marl) was 1:3.6:6.2 and that of the petroleum potential was 1:37:92.
5. The calcareous marl of Organic Facies B (even nearly AB) proved to be an excellent oil producing matter. Its average petroleum potential was 32.30 kgHC/ton of rock.

   The marl of Organic Facies BC was found to be a very good source rock, its petroleum potential was 13.04 kgHC/ton of rock.

   As the siltstone of Organic Facies D had a very low petroleum potential (0.35 kgHC/ton of rock) it was naturally a non-generative rock.

   The core Z of Organic Facies B seemed to be a good source rock. Its petroleum potential was 10.05 kgHC/ton of rock.
6. On the basis of organic facies, petroleum potential and type of kerogen two zones were distinguished in the marl of core R. These differences may reflect variations in the depositional environment. Further investigation is necessary to compare these two zones to each other.
7. The thermal degradation properties of samples from core Z and from calcareous marl of core R corresponded to those of the kerogen type II and the kerogen type I, respectively. A lower percent of the organic matter of core Z was converted to oil and a smaller quantity of bitumen developed from it than from core R. At the same time the kerogen of core R required higher activation energy for generating volatile bitumen. So, its main oil producing zone began, as well as ended temperature 50 °C below than that of the other sample.

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