Annales de la Société Géologique de Belgique. T. 110 - 1987, pp. 77-84 Mai 1987

COAL-PETROGRAPHIC INVESTIGATIONS ON SAMPLES FROM THE BOREHOLES THERMAE 2000 AND THERMAE 2002 (VALKENBURG A/D GEUL, THE NETHERLANDS)

by

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(1 figure, 2 tables and 2 plates)

1. INTRODUCTION

A few spot samples from the Santonian Aachen mud-sand and the Dinantian shales and carbonates of the boreholes Thermae 2000 and Thermae 2002 have been subject of coal-petrographic analysis. Micropetrographic descriptions of the samples are based on microscopical observations (magnification X400, oil immersion). Determination of presence of organic carbon and determination of reflectance of vitrinite and metabituminite are presented and briefly discussed.

2. MICROPETROGRAPHIC DESCRIPTIONS

2.1. THERMAE 2000

Four samples of black shale have been studied: 225-227 m, 239-242 m, 294-297 m and 324-327 m.

Sample 225-227 m consists of non-laminated to poorly laminated black shales with abundant organic matter (cf. table I). The organic matter is present mainly as primary oxidized detrital plant fragments. Vitrinite (coalified humic substance) and metabituminite (high rank bitumen) are rare. Remarkable is the presence of large sclerotioid grains (for detailed description and discussion on their nature see Bless et al. 1976). The shales contain abundant early diagenetic, framboidal pyrite suggesting a reducing depositional environment.

Sample 239-242 m has yielded black shale fragments as well as chips of partly or entirely silicified limestones. The sediment and the organic material resemble those of sample 225-227 m. The silicified rock fragments contain little or no organic matter. This explains the much lower C_{org} content of this sample as compared with the above one. Sclerotioid grains have not been observed. Along with abundant framboidal pyrite some zinc-sulfide occurs.

The rock chips of sample 294-297 m are marked by intense silicification. The black shale fragments contain oxidized detrital plant fragments, but no vitrinite. Intensely silicified rock fragments with authigenic carbonate crystals contain intergranular bitumen («Zwickelbitumen»). This means that bitumen was drived into interparticle pores by the newly formed minerals.

Sample 324-327 m has yielded a poorly laminated black shale that is partly silicified and carbonated. Thin linings of metabitumen with a fine mosaic texture occur in microfissures. This mosaic texture is produced by a variable reflectance pleochroism. Presumably the bitumen was drived off the sediment by a heating process and migrated into the microfissures. Subsequently it was converted into coke by a sudden increase of the heating.

Table I - Organic carbon in spot samples from Thermae 2000 and Thermae 2002.

THERMAE 2000

depth (m)	C_{org} (%)
225 - 227	4.31
239 - 242	1.30
294 - 297	0.63
324 - 327	1.11

THERMAE 2002

depth (m)	C_{org} (%)
182 - 184	1.33
210 - 212	3.86
228 - 230	2.33
280 - 282	1.26
311 - 313	1.90

2.2. THERMAE 2002

One sample of lignite-bearing mud-sand of the Santonian (182-184 m), and four samples of Dinantian black shale (210-212 m, 228-230 m, 280-282 m, 311-313 m) have been studied.

Sample 182-184 m is a kaolinic mud-sand of the Santonian Aachen Formation. It contains fossil charcoal (pl. 1, figures 1-2), spores and cuticules, as well as wood fragments of subbituminous rank.

Sample 210-212 m consists of finely laminated, pyrite-rich sapropelic shale. The microlamination is produced by an alternation of clay laminae and laminae with finely dispersed or concentrated metabitumen. Larger organic fragments consist of oxidized plant fragments and rare vitrinite. Large sclerotioid grains are common. Vitroplast texture has been

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PLATE 1

1-2 *Fossil charcoal derived from angiosperms.*
Kaolinic mud-sand of Santonian Aachen Formation (Hergenrath Clay equivalent).
Thermae 2002, 182-184 m.
Reflected light, oil immersion, about X140.

3-5 Sclerotioid grains of different size and structure.
Large grains (fig. 3-4) characterize the Dinantian black shales and silicified limestones in Thermae 2000 (above 268 m) and Thermae 2002 (above 250 m). Small grains (fig. 5) rarely occur also slightly deeper. The large grain in fig. 3 has been divided by shrinkage cracks. These cracks are also visible in fig. 4.
Reflected light, oil immersion, about X140.
3-4 : Thermae 2000, 225-227 m, 5 : Thermae 2002, 311-313 m.

6-7 Examples of vitrinite in Dinantian sequence, which have been used for reflectance measurements.
Thermae 2002, 228-230 m.
Reflected light, oil immersion, about X360.

8 Small vitrinite fragments in intensely silicified Dinantian black shale. Thermae 2002, 311-313 m.
Reflected light, oil immersion, X360.

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observed in one of the vitrinites, i.e. the vitrinite has become plastic along its border by heating. Vitroplast textures only develop when the rank of coalification is not higher than fat coal. The sample contains abundant organic matter (cf. table I).

Sample 228-230 m largely resembles sample 210-212 m. The organic matter is so abundant that some laminae consist of pure sapropelic coal. Sclerotioid grains are less common. One of the polished sections contains two grains which resemble petrolcoke. One of these includes so-called mesophases (pl. 2, fig. 5), spherical aggregates displaying a reflectance pleochroism similar to «Brewster crosses». These mesophases are partly fused into long chains of crystalline texture. Mesophases develop at temperatures of about 450°C. Vitroplast textures have also been observed along the borders of fine bitumen schlieren (pl. 2, fig. 4). The sample has been affected by intense mineralization. In one case (pl. 2, fig. 6) «Schalenblende» is lining a vitrinite particle.

Sample 280-282 m consists of microlaminated sapropelic shale and intensely silicified rock fragments. The shale contains abundant organic matter, whereas intergranular bitumen occurs in the silicified rock. Occasionally sclerotioid grains are present. The sample contains abundant pyrite and «Schalenblende».

Sample 311-313 m resembles petrographically sample 280-282 m. However it is more intensely silicified and carbonated. Newly formed minerals have drived the organic matter into pores and microfissures, and these have destroyed the microlamination. Only some very small sclerotioid grains occur.

Table II - Reflectance measurements on spot samples from Thermae 2000 and Thermae 2002.

THERMAE 2000

depth (m)	Vitrinite			Metabituminite		
	R_{max}%	n	s	R_r%	n	s
225 - 227	5,43	11	0,62	4,22	5	0,15
239 - 242	5,02	5	0,19	4,38	4	0,22
294 - 297	-	-	-	4,28	30	0,18
324 - 327	5,85	1	-	4,53	11	0,24

THERMAE 2002

depth (m)	Vitrinite			Metabituminite		
	R_{max}%	n	s	R_r%	n	s
182 - 184	0,47	9	0,04	-	-	-
210 - 212	5,70	10	0,37	4,33	8	0,27
228 - 230	5,78	16	0,65	4,48	10	0,24
280 - 282	5,79	6	0,40	4,12	10	0,27
311 - 313	5,76	5	0,55	4,50	15	0,30

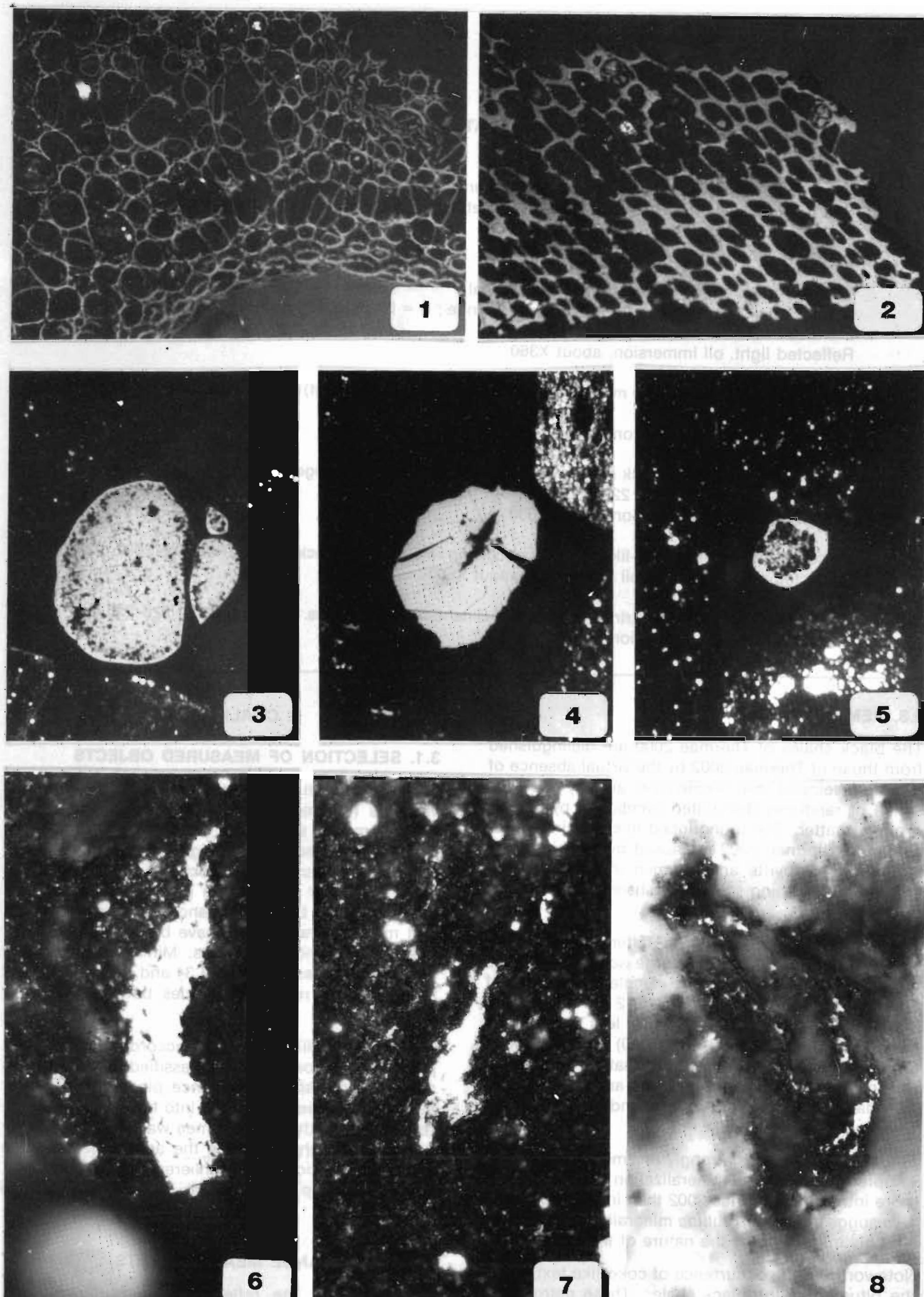
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PLATE 2

1 Sapropelic shale with microlamination, «Kupferschiefer» type.
Black = clay minerals ; greyish schlieren = metabituminite ; white spots = syngenetic pyrite. Dinantian sequence, Thermae 2002, 210-212 m.
Reflected light, oil immersion, about X360.

2 Transition sapropelic shale into sapropelic coal.
Large white spot = epigenetic pyrite ; v = vitrinite ; b = bituminite. Dinantian sequence, Thermae 2002, 228-230 m.
Reflected light, oil immersion, about X360.

3 Dinantian black shales. The microlamination (bottom left to top right) has been destroyed by silicification. Thermae 2000, 294-297 m.
Reflected light, oil immersion, about X360.

4 Vitroplast in Dinantian black shales. Contrary to vitroplast of hydrogenation residues this particle shows anisotropy. Thermae 2002, 228-230 m.
Reflected light, oil immersion, about X360.

5 Mesophases in petrolcoke-like material occurring in Dinantian black shales. Thermae 2002, 228-230 m.
Reflected polarized light, oil immersion, about X360.

6 «Schalenblende» lining vitrinite (v) grain in Dinantian black shales. Thermae 2002, 228-230 m.
Reflected light, oil immersion, about X360.

2.3. REMARKS

The black shales of Thermae 2000 are distinguished from those of Thermae 2002 by the virtual absence of a well-developed microlamination and by the presence of randomly distributed framboidal pyrite and organic matter. The pronounced microlamination of shales in Thermae 2002 is caused by the high concentrations of pyrite and bitumen in microlaminae, some of these being sapropelic shale or even sapropelic coal.

Apart from these petrographical differences there are also obvious resemblances. Large sclerotioid grains are common to abundant in sample 225-227 m of Thermae 2000 and sample 210-212 m of Thermae 2002. These grains are absent or less common in samples 239-242 m (Thermae 2000) and 228-230 m (Thermae 2002). Secondary silicification and carbonation mark the samples 294-297 m and 324-327 m of Thermae 2000, and 280-282 m and 311-313 m of Thermae 2002.

The sapropelic shales strongly resemble the Permian «Kupferschiefer». The mineralization (sulfide ores) is more intense in Thermae 2002 than in Thermae 2000. This suggests that the sulfide mineralization has been strongly influenced by the nature of the rocks.

Noteworthy is the occurrence of coke-like textures in the bitumen of the black shales. These petrolcokes indicate a rapid and presumably short-term heating when the bitumen was still mobile and not so highly coalified.

3. COALIFICATION

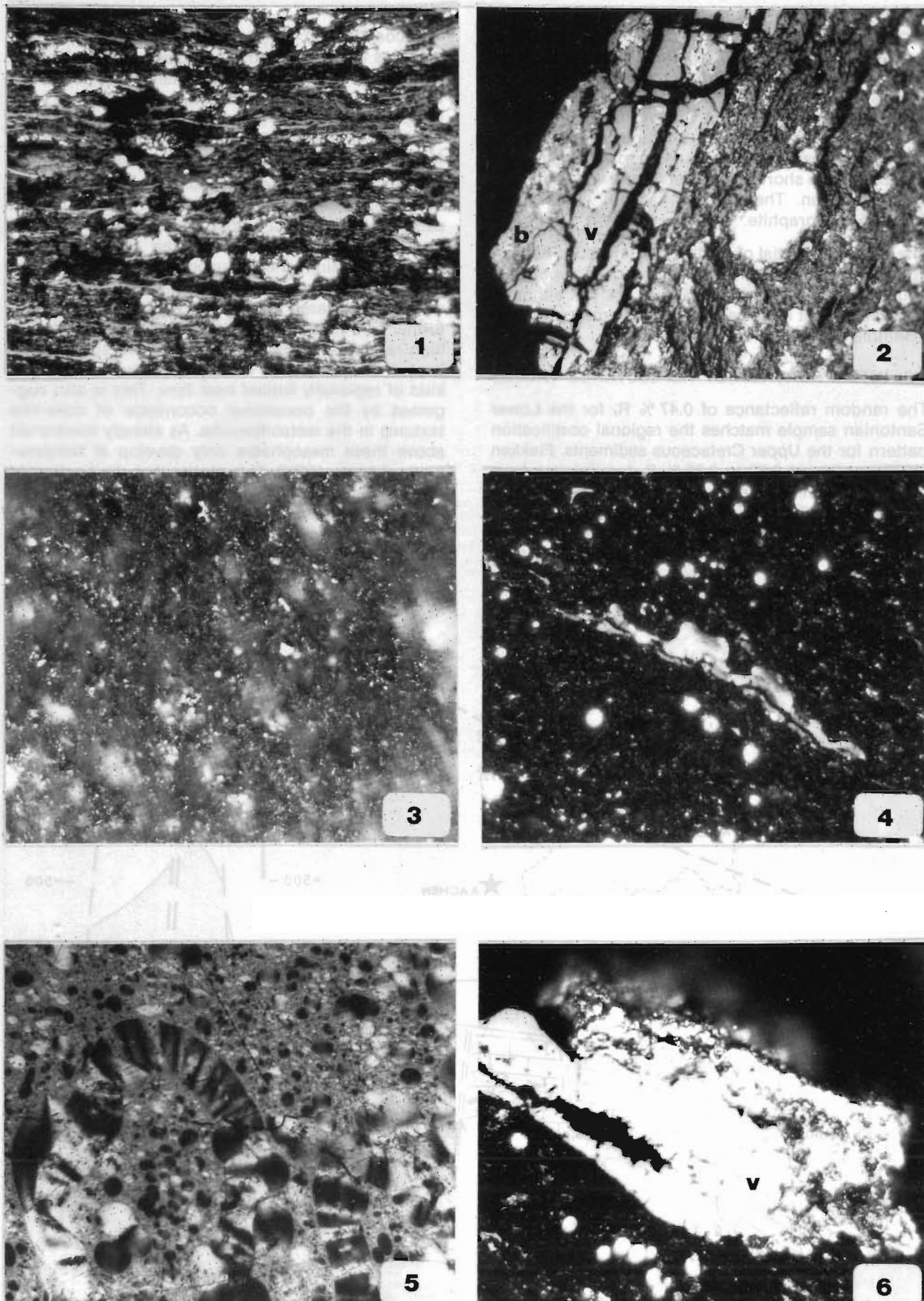
3.1. SELECTION OF MEASURED OBJECTS

As mentioned in the micropetrographic descriptions, vitrinites (the most useful macerals for reflectance measurements) are rare in all samples. Vitrinites are easily recognized in these highly coalified sediments by their pronounced bireflectance under polarized white light. For one sample (Nr. 87040, 228-230 m, Thermae 2002) the bireflectance values of maximum and minimum reflectance have been determined for the same vitrinite fragments. Minimum reflectance turned out to vary between 1.34 and 3.38 %, whereas the maximum reflectance varies between 4.62 and 6.16 %.

The highly coalified bitumen (according to the coal-petrographic nomenclature classified as metabituminite does not show reflectance pleochroism inasfar as it has not been converted into coke. Therefore the reflectance of the metabitumen was measured under unpolarized light. This has the advantage that also very small particles and schlieren can be measured, since the sample has not to be turned under the microscope.

3.2. REFLECTANCE MEASUREMENTS

The results of the reflectance measurements are shown in table II. The number of measurements is highly variable and sometimes extremely low. This makes the interpretation rather difficult. As a general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conclusion it may be stated that the coalification of the Dinantian samples is comparable for both boreholes and has reached the meta-antracite stage (> 5 % R_{max} vitrinite reflectance). The reflectance of the metabituminite is about 1 % lower than that of the vitrinite. No increase of the reflectance with the depth could be observed. This could hardly be expected because of the short depth interval and very high rank of coalification. The material has not reached the stage of semigraphite.

The organic material of the Upper Cretaceous Aachen Formation in Thermae 2002 has reached subbituminous rank (random reflectance 0.47 % R_r), although the humic plant remains under the microscope rather resemble lignite.

3.3. INTERPRETATION OF REFLECTANCE VALUES

The random reflectance of 0.47 % R_r for the Lower Santonian sample matches the regional coalification pattern for the Upper Cretaceous sediments. Flekken (1975) measured 0.32 to 0.36 % R_r for vitrinites from the Hergenrath Clay (equivalent of the here observed mud-sand) in the Cartiels borehole near Gulpen, and 0.45 to 0.46 % R_r for the same strata near Welkenraedt. This indicates that the Upper Cretaceous deposits of Valkenburg have not been affected by a geothermal anomaly.

The coalification pattern of the Dinantian rocks also matches those of nearby boreholes, notably Houthem (DB 105). In Houthem the reflectance is 4.57 % R_{max} for vitrinites from 305 m (Bless et al. 1976). This value matches rather well those for metabituminite in the Thermae boreholes. Presumably the reflectance data for Houthem were also measured on metabituminite.

The coalification of the Dinantian rocks in South-Limburg is extremely high in comparison with the coalification values for the Upper Visean (V3b) observed along the northern border of the Brabant Massif (Campine Basin), where Muchez et al. (in press) determined values of 0.92-1.18 % R_{max} along the borders of the massif and values of 2.67-2.82 % R_{max} further to the north towards the basin center. A very high rate of subsidence does not seem sufficient to explain this anomaly. There must have been some kind of regionally limited heat flow. This is also suggested by the occasional occurrence of coke-like textures in the metabituminite. As already mentioned above these mesophases only develop at temperatures of some 450°C. This means that the heating of the sediment must have been up to 450°C for a short period. The heating has partly mobilized the bitumen and converted it into coke, whereas the vitrinite was not or only superficially (vitroplasts) affected by coking process. The reflectance values of the organic matter must have been increased in the area of the

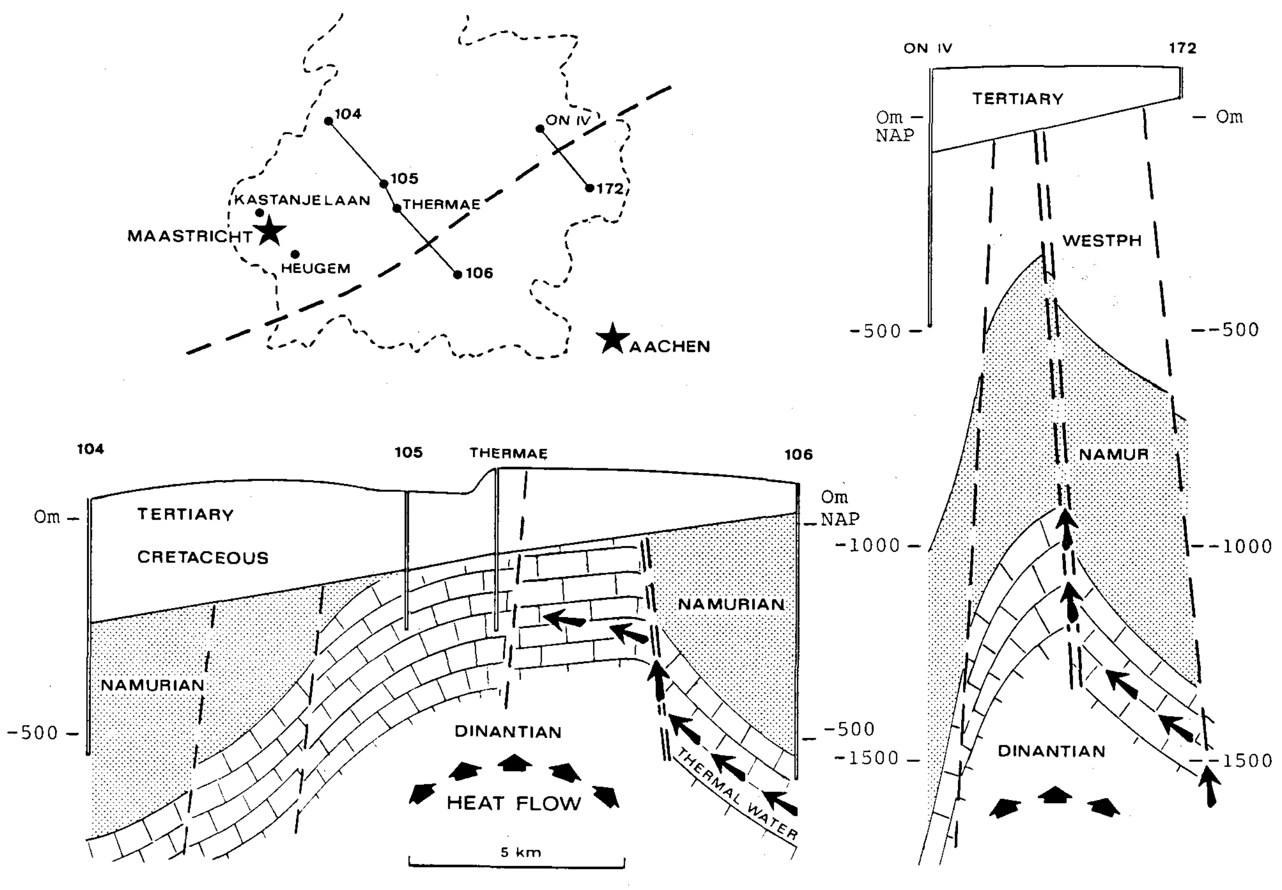

Figure 1 - Hypothetical cross-sections through the Visé-Puth Uplift (section 104-105-Thermae-106) and Waubach Uplift (section ON IV-172) showing possible position of heat flow and migration of thermal water. The heat flow is supposed to have been related with the uplift. Along with this heat flow ascended part of the mineral solutions?. The ascendant thermal water partly follows karst horizons and joints in the Dinantian carbonates and partly follows the fault system bordering the Visé-Puth and Waubach Uplifts to the south.

heat anomaly. This anomaly at least affected the area between Valkenburg (Thermae and Houthem boreholes) and Maastricht (Heugem and Kastanjelaan boreholes ; Bless et al. 1981) since everywhere extremely high reflectance values have been measured for the Dinantian strata.

Maybe the thermometamorphic process was related somehow with the sulfide mineralization. The mobilisation of the bitumen must have occurred when the coalification rank of the organic substance was still low, since the sudden heating has affected low rank organic material that had not yet been differentiated in oil and gas on the one hand and rest kerogen on the other. At that time the reflectance values must have been lower than 1.30 % R_r and presumably even lower than 1.0 % R_r. Since such low coalification ranks are still present in the Upper Visean rocks along the northern border of the Brabant Massif, it seems impossible to reconstruct the period of heating.

4. CONCLUSIONS

The coal-petrographic investigations of the Thermae samples reveal a sudden extreme increase of the temperature in the past, which has produced the high rank of coalification. This heat anomaly has no relation with the present-day thermal anomaly of the mineral water since the Upper Cretaceous sediments do not show an unusual increase of the coalification rank.

The nature of the black shales in Thermae 2000 is markedly different from that in Thermae 2002. Apparently facies changes occur within very short distances. These facies changes seem to be linked to the mineralization intensity. Mineralization is preferably linked to samples with pronounced sapropelic shale or sapropelic coal resembling Permian «Kupferschiefer».

If the mineralization is linked somehow with the heat flow that converted part of the bitumen into coke, we might conclude that the nature of the at that time still mobile organic matter in the sediment has had an important influence on the transport and enrichment of the sulfide ores, similar to that described for the Permian «Kupferschiefer».

Anomalous coalification ranks (5-7 % R_{max} for vitrinite) have also been observed for the Devono-Dinantian of Heugem and Kastanjelaan (Bless et al. 1981) and for the Namurian of the Cartiels borehole (4.1 % R_r ; Flekken 1975) near Gulpen (about 1 km northeast of Gulpen or DB106 borehole). In this context it is noticed that to the east in the former coal-mining district of South-Limburg and in the Wurm coal-mining area north of Aachen the highest coalification ranks in the Lower Westphalian seem to be linked somehow to the WSW-ENE striking fault system (Antiklinaal Fault, Oranje Fault) that forms the southern border of the Waubach Uplift. This area is also marked by frequent sulfide mineralization and by the occurrence of warm water springs in the former coal collieries (o.a. in Oranje Nassau I water of 50°C, Wilhelmina water of 38°C ; Kimpe et al. 1978) and small springs with an anomalous water temperature of about 14°C in Cadier en Keer (Fontein pool), Wittem (moat of castle Wittem) and Eysden (well of castle De Schans).

This fault system seems to be connected to the WSW with the Faille Bordière that borders the southern margin of the Brabant Massif in Belgium. The upthrown blocks always occur north of the fault system (Brabant Massif, central portion of Visé-Puth Uplift, Waubach Uplift). The throw of these faults is more than 500 m in Belgium and seems to become less pronounced in South-Limburg and Wurm area. Presumably these mark an old (pre-Caledonian?) lineament.

Most likely, a sudden heat flow along this lineament or below the Visé-Puth and Waubach Uplifts produced the intense coalification on both sides of the fault system (fig. 1). A gradually deeper burial (because of overlying Upper Carboniferous and younger deposits) would not have resulted in the formation of mesophases and vitroplast textures as observed in the Dinantian of Thermae 2002.
This sudden heat flow might be related to the uplift of the Visé-Puth and Waubach structures. It should be noticed that gravity and magnetic surveys in 1979 have not revealed any indications for a magmatic body (Bless et al. 1980).

It is supposed that mineral solutions and in part also the ascendant thermal water migrated along this fault system to the upper portions of the structural highs. Whether the mineral solutions have ascended at least in part with this hypothetical heat flow *or* have been transported by mineral water from a source in older sedimentary rocks *or* have been remobilized by the heat flow and/or by mineral water in a later phase is as yet an open-ended question. Apparently, the Thermae boreholes confront the geologist with some enigmas which require further investigations.

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