

Thermal alteration of coal in the Khasyn coalfield, Magadan region, Russia

I.E. Stukalova^{a,*}, O.V. Rusinova^b

^a Geological Institute of the Russian Academy of Sciences, Pyzhevskiy per. 7, Moscow, 119017, Russia

^b Central Research Institute of Geological Prospecting for Base and Precious Metals (TsNIGRI), Varshavskoe sh., 129-b, Moscow, 113545, Russia

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Abstract

The Early Cretaceous coal deposits of the Khasyn coalfield are intruded by Palaeogene diabase dikes. The coal has vitrinite reflectance values of 2.0–2.5% R_o , and characteristics of normal anthracite at some distance from the dikes, but at direct contact with the dike two morphological coal varieties occur: coal inclusions in the diabase dike and dispersed carbonaceous matter within the dike rock. Both types of coaly matter have properties typical of anthracites: strong anisotropy, altered internal structure and high vitrinite reflectance values ranging from 3.8 to 5.5% R_o . The X-ray diffraction measurements of the interplanar spacing $d(002)$ and the crystallite sizes L_c and L_a show rather similar values for coal inclusions in the dike and dispersed carbonaceous matter. The additional reflection at 3.37 Å, corresponding to semi-graphite admixture, occurs in the coal and carbonaceous matter inside the dike and is absent in the natural coal outside the dike.

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1. Introduction

Although the effect of magmatic intrusion on coals is a subject of important geological and practical significance, it has not been well studied. Some data concerning the mobilization of coal substances at contact with the intrusive rock and their migration through fissures are known (Podwysocki and Dutcher, 1971); however, the alteration of the intrusive rocks and the internal structure of the organic substance have not been examined in detail. Many publications describe alteration near the intrusive-coal contact at high-temperatures (e.g., Kisch and Taylor, 1966; Podwysocki

and Dutcher, 1971; Kwiecinska and Petersen, 2004; Stewart et al., 2005; and others).

This paper describes the variable degree of coal transformation within the diabase dike along with the alteration of the dike rocks itself. Thus, we made an attempt to explain how a coal seam interacted with the cooling intrusion directly at the coal-intrusive contact. In addition to local high-temperature transformations of the organic matter at the dike margins, another process was developed in the coalfield: a relatively low-temperature coal metamorphism. The superposition of relatively low-temperature hydrothermal metamorphism on products of the high-temperature thermal coal metamorphism is a new phenomenon described in this paper.

The aim of this investigation is to understand the processes involved in the thermal alteration produced by

* Corresponding author.

E-mail address: stukalova@ginras.ru (I.E. Stukalova).

the intrusion of diabase dikes into coals at the Khasyn coalfield. The study involved the determination of the nature and the specific structural properties of organic matter concentrated in coal seams and dispersed in rock affected by the dike intrusion.

2. Geological setting and structure of the Khasyn coalfield

The Khasyn coalfield occurs in the Magadan region of Northeastern Russia, in the middle stretches of the Khasyn River, 82-km north of the town of Magadan, at the confluence of the Karamken and Khasyn Creeks, where the coal-bearing sediments of the Khasyn Formation are exposed (Fig. 1). The Magadan region hosts two coal basins and few significant coalfields (Popov, 1962; Zolotov and Pekanov, 2000). The region contains high-ash coals, commonly intruded by igneous bodies with unusual contact phenomena. The Khasyn coalfield is

structurally associated with the Okhotsk–Chukotka volcanic belt, where Early Cretaceous coal deposits occur in an area of recurrent Cretaceous to Palaeogene volcanic and intrusive activity.

According to Zolotov and Pekanov (2000), from Late Jurassic to Albian times a molassic complex of continental deposits was developed in numerous basins and depressions at the foot of Okhotsk–Chukotka volcanic belt. The complex includes significant productive coal-bearing strata, commonly containing numerous coal seams forming an excellent prospect for mining.

The base of the stratigraphic coal-bearing section at the Khasyn deposit is composed of the Early Cretaceous molassic terrigene sequence with predominant andesitic to basaltic rock composition. The coal-bearing deposits of Khasyn Formation (K_1hs) are 255- to 455-m thick and are composed of sandstones, tuffs, mudstones and siltstones (Fig. 1). These rocks commonly contain high-ash coal seams. The rank of the Khasyn coals is typically

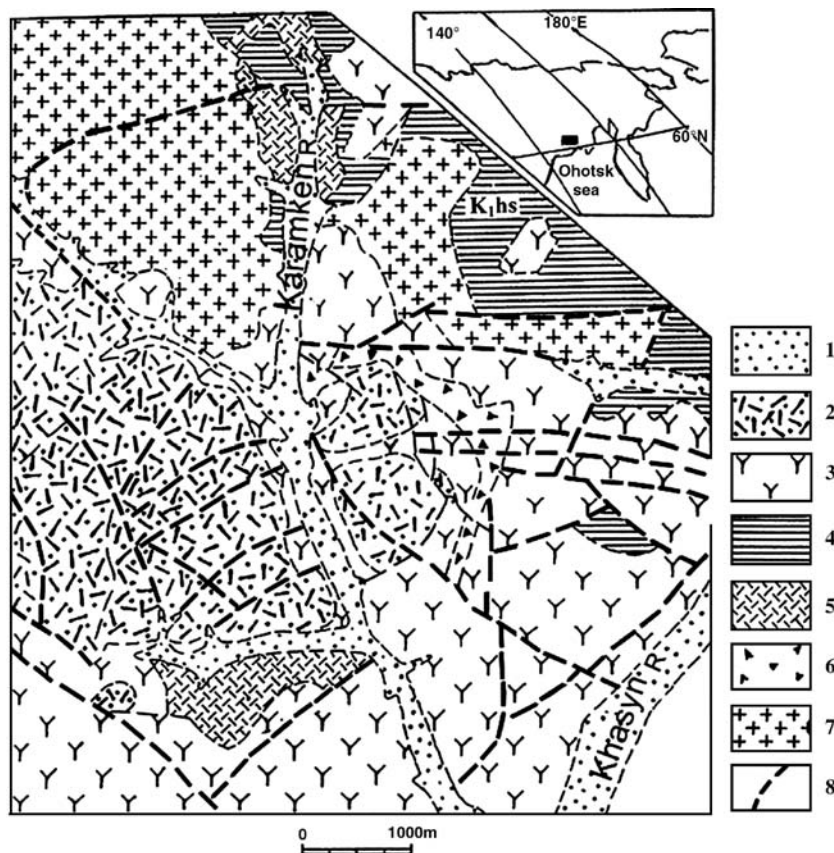


Fig. 1. Geological map of the Karamken region with the Khasyn coalfield (using field materials of A.K. Kondratenko and A.A. Krasilnikov, TsNIGRI): 1 — Quaternary alluvial sediments, 2 — Late Cretaceous Olskaya series, ignimbrites, 3 — Late Cretaceous Formation, andesites, andesite-basalts, basalts, rhyolites, mudstones, tuffs, 4 — coal-bearing Early Cretaceous Khasyn formation, sandstones, tuffs, mudstones, siltstones, coal seams (K_1hs); magmatic and volcanic rocks: 5 — Late Cretaceous subvolcanic rhyolites, 6 — Later Cretaceous automagmatic breccias, 7 — Late Cretaceous granitic rocks; 8 — faults.

between low volatile bituminous and semi-anthracite (Popov, 1962). The Khasyn Formation contains eleven coal seams with a complex structure and variable thickness, ranging from 0.6 to 5.0 m.

The Early Cretaceous coal-bearing deposits of the Khasyn Formation (K_{1hs}) in the coalfield underwent the thermal influence of the intruded Late Cretaceous acidic magmatic bodies and Palaeogene basic dikes. The latter intruding the coals are lamprophyres, basalts and diabases. At the coal-dike contact both the rock and the coal are partially or completely altered. The local high-temperature transformation of the organic matter at the dike contacts and within dikes was followed by the relatively low-temperature hydrothermal metamorphism of the coal and the dike rocks.

The Khasyn coalfield provides a good opportunity to study this subject. The coalfield demonstrates an unusual type of contact coal metamorphism, with coal seams almost totally replaced by the intrusion at contact. This process of total replacement implies that the coal burned out at high-temperatures.

The coal-bearing sequence is overlapped by the Late Cretaceous Formation (andesites, andesite–basalts, basalts, rhyolites, mudstones, and tuffs) enclosing some epithermal ore deposits; Olskaya series (ignimbrites) is the youngest in the Cretaceous volcanic sequence (Fig. 1).

The geological setting of the Khasyn deposit is characterized by the space association with younger (Late Cretaceous) volcanics with genetically related epithermal gold–silver deposits and hydrothermally altered rocks.

3. Sampling and methods

The studied samples were obtained from E.G. Peskov's collection. Peskov (1990) paid attention to some unusual relations between magmatic rocks and carbonaceous matter and found that magmatic and the host sedimentary rocks are strongly argillized near the contact with coal, he described also the aggressive behavior of the carbonaceous fluids relatively to the dike rocks (Peskov, 1990).

We studied three sample sets: (1) coal sample set (X-1) far from the dike; (2) sample set (692/M) of coal inclusions in the dike; this was divided into four sub-samples; and (3) sample set (X-2) of carbonaceous matter (CM) impregnating the dike rocks (Table 1). The dike–coal interaction is described on the base of the study of the optical and physical properties in coal and rock samples at various distances from the contact between the coal and the diabase dike.

Standard microscopic methods of coal petrography (POLAM L-213M produced by LOMO optical mechan-

Table 1
The studied samples

Sample	Characteristics	Position
X-1	Black, conchoidal fractures, anthracitic luster (a — natural sample, b — after dissolution and removal of mineral phases)	Seam coal
692/M-1	Banded, fractured coal with alternating dense bands with weak luster and loose ones with graphitic luster	Coal inclusions in the dike
692/M-2	Black homogenous fine-grained matt particles	Carbonaceous matter in the dike
692/M-3	The same as 692/M-2, but with small (around 1 mm) light spots of argillized rocks	
692/M-5	Light medium-grained undurable argillized dike rock	Argillized dike rock
X-2	Dense, fine-grained black aggregate with relics of the hydrothermally altered dike rock (around 1.5 cm) surrounded by smaller (1–3 mm) dike fragments	Carbonaceous matter in the dike

ical enterprise, St. Petersburg, Russia) with magnification 90–600 $\times$ in plane polarized transmitted and reflected light and X-ray diffraction analysis were used for detailed examination of the coals and the non-coal rocks. Vitrinite reflectances were determined with a microscope-photometer MFU-M (LOMO optical mechanical enterprise, St. Petersburg, Russia) in oil immersion with monochromatic polarized light ($\lambda=546$ nm) using LOMO standards.

The samples were also examined with a scanning electron microscope (SEM, CamScan MV2300 with an INCA 200 pad for undispensed X-ray microanalysis) using magnifications from 1000–4000 $\times$. Applying this technique the C, O, S, N contents of the coal matter were determined (the hydrogen content could not be estimated).

The X-ray diffraction patterns of powders from the rock and the carbonaceous substance were obtained by a DRON-2.0 diffractometer using Ni-filtered $CuK\alpha$ radiation, at 40 kV, 20 mA, scanning speed 1 $^\circ$ /min and recording speed 10 mm/min. The carbonaceous substance was extracted by dissolution of the mineral phases.

4. Results

4.1. Petrographic description of the dike

The dike rock is macroscopically dark gray, but in places is strongly altered and changes to light pinkish-gray colour. Under the microscope the rock (even altered) reveals a diabase structure with plagioclase phenocrysts and glassy matrix. The alteration of the diabase

was hydrothermal argillization. The plagioclase phenocrysts are replaced by clay minerals; and the matrix — by clay minerals, quartz, and carbonaceous matter. Intensive alteration concentrated around coal inclusions. In places, carbonaceous matter rims the plagioclase crystals and penetrates inside them along fissures. Also of interest is the occurrence of small bubbles enriched with the carbonaceous matter in the altered diabase (Fig. 2, photos 5 and 6). These bubbles, filled later with quartz, are probably caused by gas generation due to the thermal influence of the dike on the coal.

4.2. Petrography of organic matter

The first sample set (samples X-1a, X-1b) located far (about 10 m) from the dike display an anthracitic luster, conchoidal fractures and vitrinite reflectance values of 2.0–2.5% R_o . Polished sections examined under reflected light with magnifications of 90–400 $\times$, reveal bands of cellular plant cavities with R_{min} of 2.2% and R_{max} of 2.7% (sample X-1a).

The samples examined under the SEM demonstrated a more distinct coal structure with uncommon remains

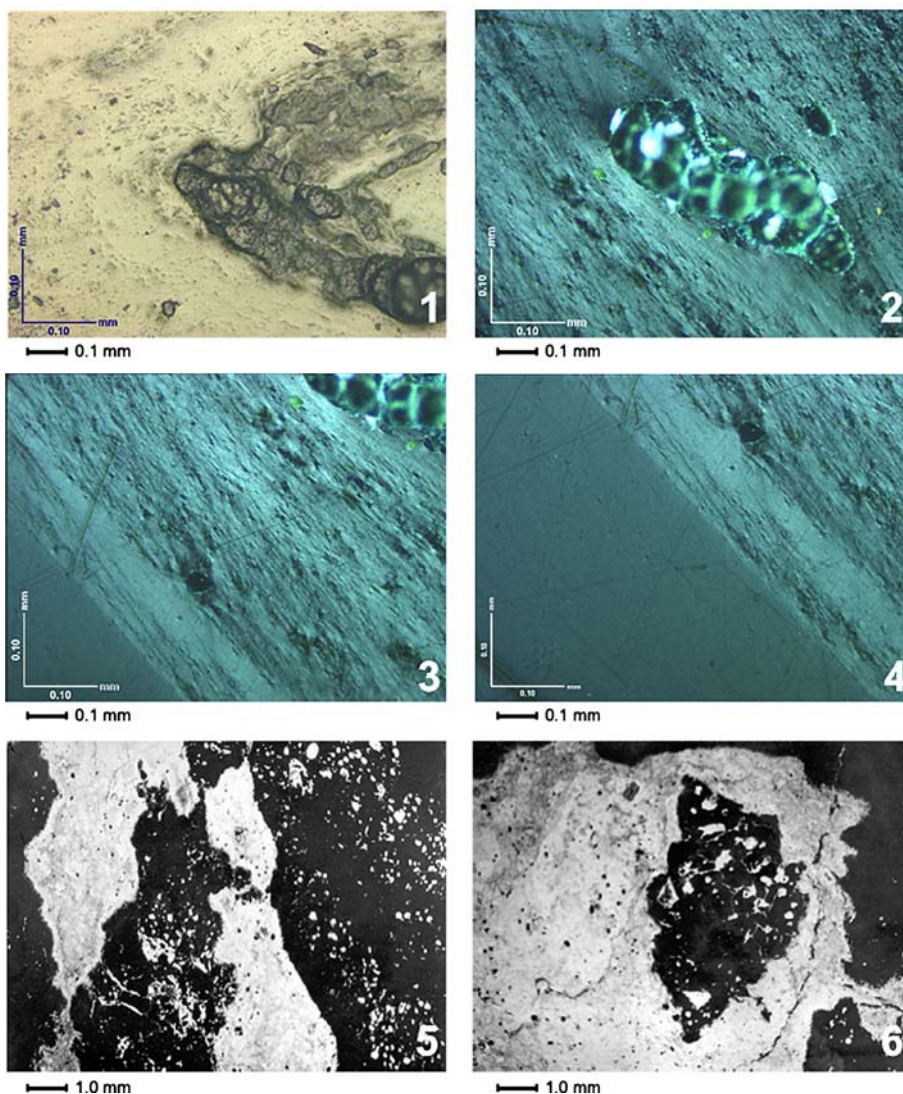


Fig. 2. Photomicrographs of the Lower Cretaceous coals and rocks in the Khasyn coalfield. 1–4 — polarized reflected light, 600 $\times$, photos 2–4 crossed polars. 1, 2 — interpenetration of organic and mineral matter fragments (sample 691/M-1); 3, 4 — two types of organic matter, inhomogeneous type with high anisotropy (photo 3) and homogeneous type with poor anisotropy (photo 4), (sample 691/M-1). 5, 6 — transmitted light, one nicol, 50 $\times$, finely dispersed carbonaceous matter with quartz inclusions in dike rock (sample 692/M-3).

of cellular plant cavities, regularly located along the bedding (Fig. 3, photos 1, 2). X-ray microanalysis combined with the SEM showed that sample X-1a contains 94.16% C, 5.10% O, 0.61% S and 0.13% Si.

The second sample set, obtained at contact with the dike, contains thermally altered coal in the form of small inclusions visible by the naked eye and having 3.8–5.5% R_o . In a typical example (sub-sample 692/M-1), coal fragments have suffered under a thermal influence typical for high-rank coal, show a metallic lustre characteristic for anthracites and strong anisotropy. Under the microscope in reflected light we can see lenticular and interbedded

carbonaceous matter, sometimes brecciated and with numerous desiccation fractures suggesting a homogeneous, and once viscous organic mass. Organic mass envelopes the rock fragments, with subsequent interpenetration of organic and mineral matter fragments (Fig. 2, photos 1, 2). Sample 692/M-1 displays 5.9% R_{max} , corresponding to the anthracite and possibly meta-anthracite stage (Stach et al., 1982; Kwiecinska and Petersen, 2004).

The coal samples in direct contact with the intrusion demonstrate a distinct banded texture with separate bands of coal fragments. The bands contain two types of organic matter: the inhomogeneous type displaying a

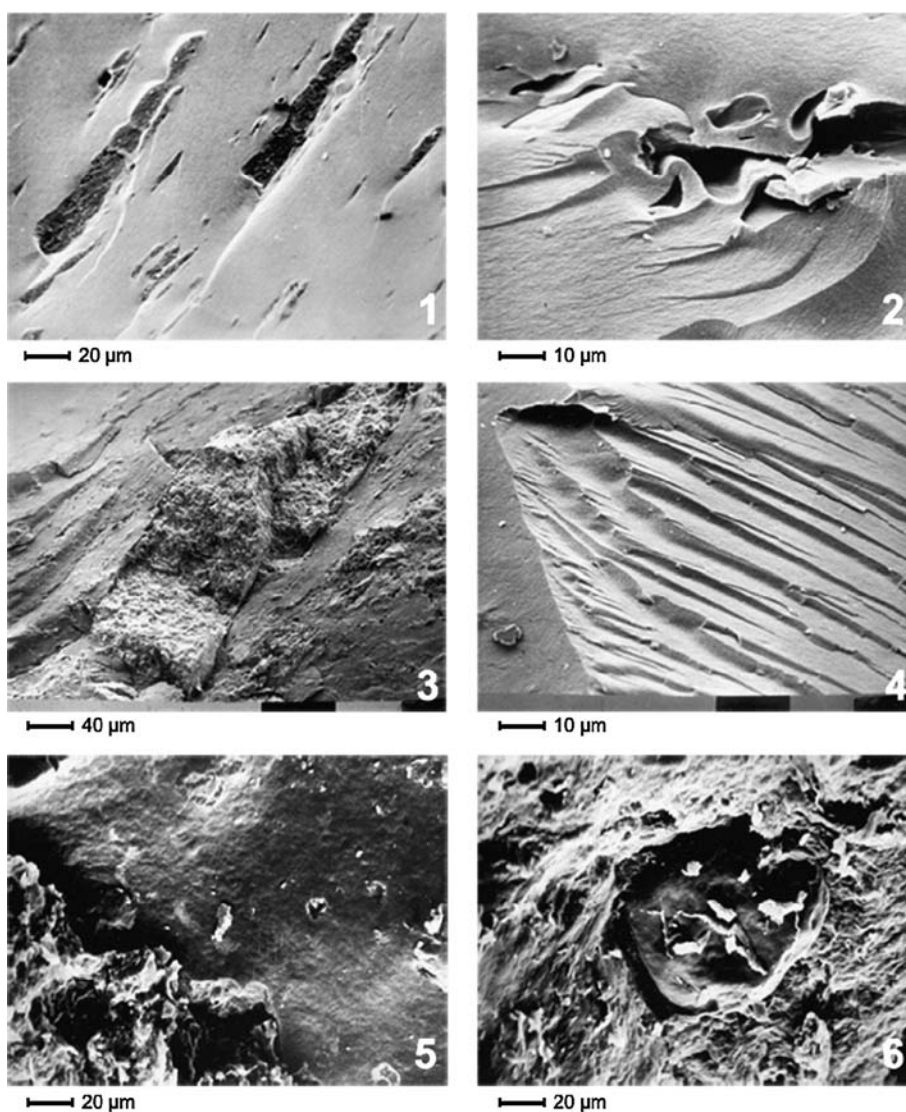


Fig. 3. Photomicrographs obtained using SEM CamScan MV2300, magnification 500–2000×. 1, 2 — uncommon remains of cellular plant cavities (sample X-1); 3, 4 — coal inclusions in the dike, the cellular structure of plant tissues is missing (sample 692/M-1); 5, 6 — Finely dispersed carbonaceous matter, unevenly distributed in the dike (sample 692/M-3).

Table 2
X-ray diffraction data and properties of the carbonaceous matter (CM)

№№ and characteristics of samples	Rock composition	Coal and CM characteristics		
		Spacing (Å)	Lc	La
X-1a Coal	CM*	15.0, 3.44, 2.10	37	23
X-1b Coal	CM**	11.6, 3.43	38	23
692/M-1 Coal in the dike	CM**, quartz, feldspar, kaoline mineral, calcite	14.7, 3.43 (+3.37)	24	33
692/M-2 Dispersed CM in the altered dike	CM**, quartz, feldspar, kaoline mineral, hydromica ($d(001)=10,3\text{Å}$)	12.6, 3.43 (+3.37)	28	20
692/M-3 Dispersed CM in the dike	quartz, feldspar, kaoline mineral, hydromica ($d(001)=10,3\text{Å}$), smectite ($d(001)=11,2\text{Å}$), admixture of CM			
692/M-5 Dike altered	calcite, quartz, feldspar, hydromica ($d(001)=10,4\text{Å}$)			
X-2 Dispersed CM in the dike	CM**, quartz, chlorite, feldspar	13.8, 3.43	21	22

*Possible occurrence of insignificant quartz, siderite, mineral of kaolinite group (total part of mineral phases to 2%), **Carbonaceous matter is studied after dissolution of mineral phases.

strong anisotropy (Fig. 2, photo 3) and the homogeneous type, which has a weak anisotropy (Fig. 2, photo 4). The inhomogeneous bands contain remains of cellular cavities filled with mineral matter. These types alternate in sharply defined bands (Fig. 2, photos 3, 4). In polarized light, especially under partly crossed nicols, the former micropetrographic heterogeneity becomes visible. In contrast to the only slightly altered coaly matter located far from the dike (2.0–2.5% R_o), the coal located in direct contact with the dike demonstrates properties typical for anthracite: high anisotropy, internal structure changes and high values of vitrinite reflectance (3.8–5.5% R_o). $R_{\min}$ in oil is 4.2%, $R_{\max}$ 5.9% (sample 692/M-1). Specific inhomogeneous texture of carbonaceous matter can be distinctly seen under crossed polarized light (Fig. 2, photo 3). The coal inclusions in the dike are more strongly metamorphosed. It becomes practically impossible to identify the nature of the original plant tissues (Fig. 3, photos 3, 4). The cellular structure of plant tissues is missing, and any microcomponents, typical for coal, are practically indistinguishable.

The third sample set of carbonaceous matter dispersed in the dike rocks (samples X-2, 692/M-2, 692/M-3, 692/M-5; see Table 1), differs in carbon content (C_{org}). Finely dispersed carbonaceous matter is unevenly distributed in the dyke: the rock is partly light-

coloured (sample 692/M-5), partly coloured black due to the carbonaceous matter (samples 692/M-2 and 692/M-3). In the latter case the carbon content (C_{org}) reaches values up to 50% (Fig. 2, photos 5, 6). $R_{\min}$ in oil is 3.9%, $R_{\max}$ 4.7% (sample 692/M-2).

It must be noted that it is rather difficult to determine the structure of transformed organic matter products in the course of its interaction with the dike, since the primary genetic features of coal (cellular texture, microcomponents) are practically missing. The structure of finely dispersed carbonaceous matter cannot be detected, even under SEM (Fig. 3, photos 5, 6).

4.3. X-ray diffraction

X-ray diffraction (XRD) was used to identify mineral phases and to obtain X-ray diffraction parameters of the coal and carbonaceous matter (CM) (Table 2). X-ray diffraction patterns (Fig. 4) were obtained for three sample sets: coal sampling far from the dike (sample X-1; a — natural, b — after dissolution of mineral phases), coal sampling in the dike (692/M-1) and CM separated from the dike rock (692/M-2, X-2).

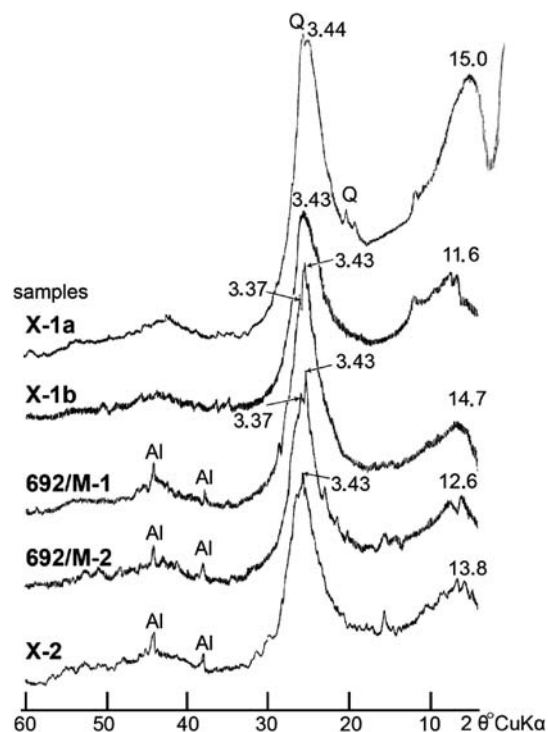


Fig. 4. Powder X-ray diffraction patterns of the studied samples: Samples X-1a, X-1b — coal; sample 692/M-1 — coal inclusion in the dike; samples 692/M-2, X-2 — dispersed carbonaceous matter in the dike. Q — quartz (4.20 Å and 3.34 Å); Al — aluminium of the specimen holder (2.37 Å and 2.05 Å).

There are three X-ray diffraction parameters reflecting the crystalline order in coals of various rank: $d(002)$ — interlayer spacing; the size of crystallites, i.e. La — average diameter; and Lc — ‘height’ of aromatic stack along c axis or the crystallite thickness (Kwiecinska, 1980; Tagiri, 1981; Stach et al., 1982). Comparison of the X-ray diffraction data ($d(002)$, Lc and La) permits identification of the structural order and the degree of graphitization of the organic matter in the studied samples.

The coals far from the dike (samples X-1a, X-1b) contain not more than 2% of mineral admixtures and provide a distinct diffraction pattern without refinement. Unaltered coal (sample X-1a) displays high peak at $d(002)=3.44$ Å, a weak indistinct peak at 2.10 Å and low-angle well developed but rather broad peak at 15.0 Å (Fig. 4). The reflections of three-dimensional ordering are absent. The size of coherent dispersion area along the c axis (Lc) is 37 Å, the average layer diameter (La) is 23 Å (Table 2). After sterling and acid treatment (sample X-1b), the low-angle peak shifts and becomes lower and wider (Fig. 4).

The diffraction pattern of coaly inclusions in the dike after removal of the mineral phases (sample 692/M-1) is similar as a whole to the diffraction pattern of natural coal. The main differences are the drift of the low-angle peak to a higher angle and the diminution of its intensity. La value changes to 33 Å, Lc is 24 Å. The reflection at $d(002)$ is equal 3.43 Å and has an additional (complicating) peak at 3.37 Å (Fig. 4).

The X-ray diffraction pattern for the third sample set after removal of the mineral phases (samples 692/M-2 and X-2) shows a main reflection at $d(002)=3.43$ Å and the low-angle reflections (12.6 Å, 13.8 Å). La are 20 Å and 22 Å, Lc are 28 Å and 21 Å, respectively.

Evaluating the X-ray diffraction data obtained from all of the studied samples, we conclude that all diffraction patterns (Fig. 4) contain the peak corresponding to $d(002)$ of anthracites, a wide peak at low-angle interval and a weakly developed peak in the interval 38–48° 2θ CuK α . Reflections $d(002)$ of all samples are strong, rather wide, and the spacings of (002) reflection are similar (3.44–3.43 Å). The carbonaceous matter and the coal in the dike have an additional peak at 3.37 Å corresponding to semi-graphite admixture. This is important, because this fact underlines the alternation of internal coal matter and the local high-temperature transformation of the organic matter at the dike contacts and within the dikes. This local high-temperature transformation was followed by the relatively low-temperature hydrothermal metamorphism of both the coal and the dike rocks.

The studied samples are identified as anthracites after calculation of crystallite size Lc, La, and $d(002)$ values

(Table 2), (according to Kwiecinska, 1980). Very low values Lc and La parameters and $d(002)=3.43$ –3.44 Å show that coal and CM are characterized by the turbostratic structure constructed by no order stacking planar layers of aromatic hydrocarbon.

The samples show similar $d(002)$ — Lc parameters commonly characterizing the low crystallinity degree; however, some differences occur. Altered coal and carbonaceous matter display a lower and wider low-angle peak than natural coal. A wide peak at 5–8° 2θ CuK α corresponds to the specific internal structure of coals with a high gas generation potential. The reflection is better developed in the unaffected coal, but in other samples it migrates toward high angles and becomes broader and lower. This probably relates to an incomplete degassing of coal.

To estimate the formation temperature of mineral assemblages associated with the carbonaceous matter transformation and the interaction of the coaly mass and the rock, we used the extent of mica hydration. The extent of mica hydration and the structural modification of the hydrated micas, determined from X-ray diffraction characteristics, reflect the temperature conditions of the formation of the mineral assemblages containing mica and mica-like minerals, following the work of White and Hedenquist (1995) on micaceous mineral thermometry. The hydromicas, replacing the dike rock, reveal a similar value of the first basal reflection $d(001)$ (Table 2), the latter providing the upper temperature limit for their formation at 200 °C. The occurrence of smectite in sample 692/M-3 indicates that the lower temperature limit for the layered silicates formation was locally below 160 °C.

5. Discussion

Many publications describe in detail the metamorphism phenomena and alteration of coal close to the intrusive-coal contact at high-temperatures (Kisch and Taylor, 1966; Podwysowski and Dutcher, 1971; Kwiecinska and Petersen, 2004; Stewart et al., 2005 and others). Kisch and Taylor (1966) paid attention to the intrusive rock alteration and indicated that the ferromagnesian minerals of the diabase dike rock are pseudomorphically replaced by carbonates derived by reaction with CO₂, which was produced by oxidation of the carbonaceous matter of the coal. In the Khasyn deposit, plagioclase in the diabase dike is pseudomorphically replaced by clay minerals.

Kisch and Taylor (1966) describe coal converted to a natural coke during a high-temperature (about 760 °C) alteration process with vitrinite becoming isotropic and

getting mosaic structure. Kwiecinska and Petersen (2004) underline that natural coke is formed from coal by a relatively local elevated heat flow caused by an intrusive body. We made an attempt to explain how a coal seam interacts with an intrusion and vitrinite became more anisotropic.

Podwysocki and Dutcher (1971) described a coal contact with lamprophyre sills. Petrographic analysis indicated a decrease in coal metamorphism degree with increasing distance from the sill, the major indicator being the maximum reflectance values varying from 1.2% farthest from the sill to 2.2% within the sill. In this paper we also underline the thermal influence of diabase dike on coal alteration and amplification of rank and anisotropy of anthracite.

We cannot capture all aspects of thermal alterations of coals at contact to intrusive rocks. However, the object of the paper is dedicated to our own experimental results of estimation of the cooling intrusion influence on carbonaceous material. The problem was to determine the nature of dispersed carbonaceous matter in rocks with $C_{org} = 1\text{--}2\%$. We cannot measure the vitrinite reflectance of this material. And we cannot determine its nature, i.e. if this material is humic or not. The important result is that X-ray diffraction characteristics of concentrated and dispersed carbonaceous matter appears to be the same. The finely dispersed carbonaceous matter is the result of destruction and re-deposition of coal matter.

The results of our study allow to conclude that at the early stage of the coal alteration the temperature was not so high as in the case described in Kisch and Taylor (1966) and others. This stage was followed by a rather low-temperature alteration process. The accompanying alteration of the dike rock relates to the hydrothermal argillization at 160–200 °C. Probably, some structural canals for fluid convection occur in the dike that complicated the dike–coal relationships.

The injection of the carbonaceous matter into the dike was accompanied by the degassing, the gas bubbles formation, and later on, by the quartz deposition.

The carbonaceous matter of hydrothermal breccias in the gold–silver epithermal deposits that are common in the study region (besides coalfields) probably reveals the same nature as the disseminated carbonaceous matter in the studied dikes. The presented data supply additional information concerning the behavior of the carbonaceous matter in hydrothermal processes, not only within black shales (Rusinova et al., 1996), but also in various epithermal deposits.

6. Conclusions

1. The local high-temperature transformation of the organic matter at the dike margins was followed by the relatively low-temperature hydrothermal metamorphism. At the coal-dike contact both the rock and the coal are partially or completely altered.
2. Based on calculations of crystallite size L_c and L_a and the $d(002)$ values, the altered coal and carbonaceous matter are identified as anthracite.
3. The X-ray diffraction characteristics of concentrated and dispersed carbonaceous matter appear to be similar as a whole, but show some differences. The studied samples have similar $d(002) - L_c$ parameters that commonly characterize the crystallinity degree, but the altered coal and the carbonaceous matter reveal lower and wider low-angle reflection than the natural coal. This difference probably relates to partial degassing of coal.
4. The carbonaceous matter and the coal in the dike have an additional reflection at 3.37 Å corresponding to semi-graphite admixture.
5. The alteration of the diabase relates to the hydrothermal argillization. Probably, the dike was saturated with structural canals for fluid convection that complicated the dike–coal relationships. The diabase rich in carbonaceous matter contains gas bubbles filled later with quartz. Perhaps these bubbles resulted from gas generated due to the thermal influence of the dike on the coal.

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