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Lignite oxidation under the influence of glacially derived groundwater: The pyropissite deposits of Zeitz-Weißenfels (Germany)☆

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ABSTRACT

The Middle Eocene lignites of the Zeitz-Weißenfels coal mining area in central Germany hosted one of the most economically successful lithotypes of lignite deposits utilised in the paraffin industry of the 19th century, i.e. the pyropissite deposits. However, due to their economic significance, these rare lithotypes were almost completely mined out, such that presently only a few remnant deposits are known. Apart from the Zeitz-Weißenfels coal mining district, other pyropissite deposits were also encountered in other lignite mines, for instance, in the Hessian and Subhercynian Basins.

Pyropissite is a whitish and bitumen-rich variety of soft brown coal (lignite) lithotypes, dominated by a homogeneous matrix with loosely embedded organic and inorganic components. Fresh exposure of pyropissite at Grana was logged and studied petrographically and geochemically, along with samples obtained from archival collections. Results were compared with those from the 19th and 20th century to derive general conclusions regarding origin and formation of pyropissite.

Microscopically, the main constituent of this special lignite lithotype is amorphous to detrital in appearance. According to W. Schneider the term xanthinite is applied to this component, which forms the groundmass of the pyropissite. It contains high proportions of liptinitic substances with contributions of formless huminitic material and minor amounts of mineral matter. Geochemically, pyropissite is characterised by an increased H/C atomic ratio and a very high content of bitumen (in terms of toluene-soluble components) as well as low-temperature carbonisation tar.

The obtained data revealed that the extraordinarily high content of liptinite and the paucity of huminitic material of the pyropissite deposits of the Zeitz-Weißenfels coal mining area are not related to primary depositional processes but rather are the result of dissipation of high condensed huminites and the consequent enrichment of liptinitic substances. The mechanism involves oxidation by groundwater and is related to glaciation processes operating during the Pleistocene. Such glacially derived processes also account for both vertical and horizontal distributions of the pyropissite.

Thus, recent research activities may also serve as an innovative inspiration to fundamental understanding of the formation of lignites with extremely high liptinite content.

1. Introduction

The general geology, petrography and stratigraphy of the Eocene lignite deposits of the Leipzig Embayment (formerly called "Weißelster Basin"), has been thoroughly investigated for > 200 years due to their high economic significance. Therefore, this region ranks among the

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most explored lignite deposits of the world (e.g. Eissmann, 2002a; Standke et al., 2002; Rascher et al., 2008; Standke, 2011).

Apart from its economic worth, the paraffin industry of the 19th and 20th centuries were particularly interested in the localised outcrops of pyropissite (i.e. special lithotype of soft brown coals). These pyropissite deposits were located in the southwestern part of the basin – between the cities Zeitz and Weißenfels.

The pyropissite occurs as a firm and massive layer at the top of the main seam of the central German lignite basin (i.e. lignite seam 23), with a thickness ranging from some decimetres up to two meters. Fresh pyropissite sample is brownish in colour wirth greasy to plastic properties. The colour of the dry sample is observed to be yellowish to whitish and its consistence becomes brittle and crumbly. From a chemical point of view, pyropissite exhibits hydrophobic properties and a low specific gravity, enabling it to float in water even in a fresh condition testifying to its positive buoyant property. Furthermore, dried pyropissite is highly flammable and burns with a lucid but strongly sooting flame, dripping burning drops almost like sealing wax (Voigt, 1799). Above all, its specific chemical composition characterised by high hydrogen contents, an extraordinary high content of extractable bitumen and up to 40 wt.-% (daf) of carbonisation tar (Teichmüller, 1982) explains its preferred application in low-temperature carbonisation (Table 1).

Due to its high bitumen and tar yields (Table 1), pyropissite was of high economic value, especially for the chemical industry of the 19th century. However, the worth of this raw material was not recognised in the mining area of Zeitz-Weißenfels for relatively long time. Voigt (1799), who described a waxy coal from Helbra (Saxony-Anhalt, Germany), first referenced it. However, in the region of Grana even in the 1830s pyropissite was thought to be a clayey mud because of its massive habitus. Just by accident, its real nature as a coal deposit was discovered in this region in the early 1840s. Consequently, pyropissite was classified as an immature lignite lithotype and used together with conventional brown lignite lithotypes for producing coal briquettes. Their high combustibility made them well known and being requested in the nearby city Leipzig for the purpose of civil fuel material (Bellmann et al., 2017). In 1845, Wackenroder and Staffel conducted a first scientific research of pyropissite, followed by many others. As a result, between 1850 and the 1880s a thriving low-temperature carbonisation industry developed based on the pyropissite deposits. However, due to the intensive open pit and underground mining activity, this rare and sparse lignite lithotype was almost completely worked out, causing cessation of its related chemical industry. As a result, just a few scientists researched the residual and uneconomic pyropissite occurrences at the early beginning of the 20th century. In the following decades, the subject of pyropissite and its industrial applications sank almost into oblivion.

Table 1
Comparison of soft brown coal lithotypes of the Leipzig Embayment (brown, yellow, pyropissite) regarding their chemical composition (compiled after Karsten, 1850; Bischof, 1850; Grotowsky, 1876; Boltze, 1877; Schwarz, 1879; Riebeck, 1880; Raefler, 1912; Vulpius, 2015).

	Soft brown coal lithotypes of the Leipzig Embayment		
	Brown lithotype	Yellow lithotype	Pyropissite
Proximate analysis [wt.-%]			
Raw coal water content W	55.0–63.0	55.0–63.0	60.0–68.0
Ash content A^{d}	8.0–16.0	12.0–16.0	5.0–15.7
Fixed carbon $C_{fix}{}^{daf}$	41.0–44.0	35.0–38.0	< 10.0
Volatiles V^{daf}	56.0–59.0	61.0–64.0	> 90.0
Ultimate analysis [wt.-%]			
Carbon C^{daf}	~64.0	~70.0	~74.0
Hydrogen H^{daf}	~5.0	~6.0	~11.0
Oxygen O^{daf}	~25.0	~18.0	~14.0
Nitrogen N^{daf}	0.3–0.5	0.5–0.6	0.2–0.3
Sulphur, total $S_t{}^{daf}$	8.0–9.0	~7.0	1.0–1.5
Atomic ratios [−]			
H/C	0.80–0.90	~ 1.00	1.60–2.00
O/C	0.20–0.30	0.15–0.20	0.11–0.14
Economic yields [wt.-%]			
Bitumen (toluene-soluble part) B^{daf}	2.2–7.1	16.1–29.8	25.0–58.3
Low-temperature carbonisation tar T^{d}	7.5–10.7	8.6–14.3	30.0–40.0
Specific particle density ρ [g/cm^3]	1.5	1.4	0.9–1.12

Research of the origin of pyropissite looks also back on a turbulent history, when a wide range of hypothesis was discussed. The first one postulated by J. Voigt (1799) assumed a colluvial origin of pyropissite by mineralisation of drifted trees in terrestrial lakes. According to Brückner (1852) pyropissite is the product of the conversion of plant relicts to resins or amber in a low-salt marine environment. Grotowsky (1876) published the opinion that pyropissite results from a natural low-temperature distillation of resin-rich woods, especially of conifers. According to him, the brownish lignites underneath the pyropissite represent the remaining carbon.

At the turn of the 20th century, the general discussion about the allochthonous or autochthonous formation of lignite also considered questions of the origin of pyropissite: Von Fritsch (1890) as well as Tille (1915) was of the opinion that lignite and pyropissite result from the allochthonous accumulation of plant debris, followed by density segregation. Hence, the accumulation of lignite takes place first, followed by the lighter resin-like components of the pyropissite layer at the top. The same mechanisms were considered by Witt (1902) and Kraemer and Spilker (1902), but assuming different starting materials. Witt (1902) suggested accumulations of spores from huge fern forests while Kraemer and Spilker (1902) presumed green algae to accumulate during several thousands of years. Potonié (1908, 1910) concluded an intermediate theory of a primary autochthonous coal formation, followed by a secondary transport of the uppermost layers and their re-sedimentation depending of their density. Herter (1858), Fiebelkorn (1895), Heinhold (1905, 1906), Hübner (1906) and Raefler (1912, 1920) suggested the autochthonous origin of pyropissite but with its continuous in-situ carbonisation.

In addition to these major contributions, many smaller ones discussed whether the different lithotypes are created

(1) by different plant associations colonising the bog depending of environmental conditions or
(2) by selective decay of different plant tissues in the peat stage with the early diagenetic huminitic material being decomposed under aerobic conditions with consequent enrichment of the lipid- and resin-rich plant materials.

In both cases, pyropissite is believed to represent an extreme variety of the yellow and bitumen-rich lignite lithotype of the Leipzig Embayment (so called "Schwelkohlen") – formed either due to a very special and lipoid-rich plant association at the margin of the ecosystem or by intensive aerobic destruction of the plant material caused by unique environmental conditions. The latter scenario was also preferred by M. Teichmüller (1982) as one of the last scientific researchers working with pyropissite.

At present, a clay open pit mine located near Grana in the northwest of Zeitz, Germany, offers a new access to such a residual pyropissite occurrence providing a rare possibility for pyropissite examination (Bellmann et al., 2017). This pyropissite profile was compared to a standard lignite profile from lignite seam 23 at the open pit mine at Profen and to historic sample material. This research serves as the foundation for reviewing and expanding the former knowledge of pyropissite. It also plays a tribute to Marlies Teichmüller's scientific study on the legendary pyropissite.